Predicting De Facto Reuse Impacts on Drinking Water Sources

at Small Public Water Systems

by

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ABSTRACT

De facto potable reuse (DFR) occurs when surface water sources at drinking water treatment plants (DWTPs) contain treated effluents from upstream wastewater treatment plants (WWTPs). Contaminants of emerging concerns (CECs) originate from treated effluents (e.g., unregulated disinfection by-products, pathogenic microorganisms as *Cryptosporidium* oocyst, *Giardia* cyst, and *Norovirus*) can be present in surface water and pose human health risks linked to CECs. Previously developed De facto Reuse Incidence in our Nations Consumable Supply (DRINCS) model predicted DFR for the national largest DWTPs that serve >10,000 people (N = 2,056 SW intakes at 1,210 DWTPs). The dissertation aims to quantify DFR at all surface water intakes for smaller DWTPs serving ≤10,000 people across the United States and develop a programmed ArcGIS tool for proximity analysis between upstream WWTPs and DWTPs. The tested hypothesis is whether DWTPs serving ≤10,000 people are more likely to be impacted by DFR than larger systems serving > 10,000 people.

The original DRINCS model was expanded to include all smaller DWTPs (N = 6,045 SW intakes at 3,984 DWTPs) in the U.S. First, results for Texas predicted that two-thirds of all SW intakes were impacted by at least one WWTP upstream. The level of DFR at SW intakes in Texas ranged between 1% to 20% under average flow and exceeded 90% during mild droughts. Smaller DWTPs in Texas had a higher frequency of DFR than larger systems while < 10% of these DWTPs employed advanced technology (AT) capable of removing CECs. Second, nationally over 40% of surface water intakes at all DWTPs were impacted by DFR under average flow (2,917 of 6,826). Smaller DWTPs had a higher frequency (1,504 and 1,413, respectively) of being impacted by upstream

WWTP discharges than larger DWTPs. Third, the difference in DFR levels at smaller versus larger DWTPs was statistically unclear (t-test, $p = 0.274$). Smaller communities could have high risks to CECs as they rely on surface water from lower-order streams impacted by DFR. Furthermore, smaller DWTPs lack more than twice as advanced unit processes as larger DWTPs with 52.1% and 23%, respectively. DFR levels for DWTPs serving > 10,000 people were statistically higher on mid-size order streams (3, 5, and 8) than those for smaller DWTPs. Finally, DWTPs serving > 10,000 people could pose risks to a population impacted by DFR > 1% as 40 times as those served by smaller DWTPs with 71 million and 1.7 million people, respectively. The total exposed population to risks of CECs served by DWTPs impacted by upstream WWTP discharges (DFR >10%) was estimated at 12.3 million people in the United States. Future studies can use DRINCS results to conduct an epidemiological risk assessment for impacted communities and identify communities that would benefit from advanced technology to remove CECs.

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"What is your name?" "My name is Thủy and it means "Water" in Vietnamese. And that is my destiny to study Water". That is the best answer that I would like to say again and again. My name was given to me by my beloved grandfather and reflects my passion for drinking water quality in my life.

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CHAPTER 1

INTRODUCTION

Municipal wastewater effluents that discharge into inland surface water (i.e., rivers, lakes, reservoirs, canals, etc.) play a critical part in the total available water resources in the United States (Reuse, 2012). Over 16,000 WWTPs are discharging 33 billion gallons of treated wastewater daily, and 23.7 billion gallons per day are discharged directly into surface water sources (USEPA, 2012). Surface water sources account for more than 60% of the nation's water withdrawal and provide public supply for more than 87% of the total U.S. population (or 283 million Americans) (Dieter et al., 2018). The U.S. Environmental Protection Agency (EPA) categorized public water systems based on their population served. Their five sizes are enlisted as: Very Small systems serve less than 500; Small systems, serve 501-3,300 people; Medium systems, serve 3,301 – 10,000 people; Large systems, serve 10,001 – 100,000, and Very Large systems serve more than 100,000 people (USEPA, 2019a). The Safe Drinking Water Act (SDWA), under Title 40 of the Code of Federal Regulations (CFR) Part 141, was passed by Congress in 1974 and amended in 1996 (USEPA, 1996b). Under the SDWA, the U.S. EPA established the National Primary Drinking Water Standards and set quantitative values on Maximum Contaminant Levels (MCL) and Maximum Residual Disinfectant Level (MRDL) for more than 90 contaminants in drinking water to ensure safe and clean water for public supply (USEPA, 1996a). The U.S. EPA also established Treatment Techniques Requirements (TTR) to control limit levels of some contaminants (e.g., viruses, bacteria, and turbidity) at public water systems. Among other types of drinking water violations, health-related violations, including any violations of MCL, MRDL, or

TTR at a public water system, can pose potential risks to public health (Kohli, Rahman, & Stavang, 2016; USEPA, 2016). Recent studies on a statistical analysis of SDWA enforcement and violations at all sized PWSs indicated that smaller PWSs (serving 10,000 or fewer people) tend to have more health-related violations than larger PWSs (serving more than 10,000 people) (Allaire, Wu, & Lall, 2018; Kirchhoff, Flagg, Zhuang, & Utemuratov, 2019; Kohli et al., 2016; Konisky & Teodoro, 2016; Oxenford & Barrett, 2016; Rahman, Kohli, Megdal, Aradhyula, & Moxley, 2010; Rubin, 2013; Switzer & Teodoro, 2018; Wallsten & Kosec, 2008). A national assessment on all sized PWSs by Wallsten and Kosec (2008) indicated that more MCL violations were likely to be committed by smaller water systems more than large water systems as large water systems may have a greater capacity to meet drinking water regulatory compliance.

Smaller public water systems (serving ≤ 10,000 people) face more challenges in providing “water of adequate quality and quantity” as they generally have less technical, management, and financial capacity than larger water systems (NRC, 1997; USGAO, 1990). The financial constraints of a smaller population served can limit the upgrade of water facilities due to a limit on the number of hired managers and staff to operate the systems. In comparison to larger public water systems, the smaller water systems have been equipped with less extensive unit processes capable of removing a broad range of contaminants of emerging concerns (CECs) (USEPA, 2011). Furthermore, smaller public water systems often struggle to maintain the same compliance with the SDWA standards (e.g., compliance schedules, standards, and monitoring requirements) as the larger water systems (NRC, 1997). Therefore, the risks of smaller water systems associated with CECs in drinking water sources may be more significant than larger water systems.

De facto potable reuse is defined as the incidental occurrence of upstream treated effluents into a public supply source (Reuse, 2012). Thus, surface water sources downstream are likely to contain wastewater-derived contaminants from WWTP point sources. There is a growing concern on several pathogenic organisms (e.g., the oocysts of *Cryptosporidium* parvum, the cysts of *Giardia* lamblia, and enteric viruses) that are highly detected in treated wastewater and resistant to chlorination. Chlorination is a conventional disinfection process and it is commonly employed at most public water systems together with other processes such as chemical coagulation, flocculation, sedimentation, and filtration through granular media (Howe, Hand, Crittenden, Trussell, & Tchobanoglous, 2012). Still rare, direct potable reuse systems use wastewater treated by a conventional wastewater treatment (primary sedimentation and biological treatment with activated sludge), followed by an advanced treatment process (microfiltration, ultraviolet advanced oxidation process, ozone, biological activated carbon, membrane bioreactors, or nanofiltration).

Quantitative microbial risk assessment (QMRA) method can be used to predict the human health risks linked to exposure to waterborne pathogens in drinking water sources (Haas, Rose, & Gerba, 1999; Reuse, 2012). The EPA guidelines indicate the log inactivation values of 12–10–10 for enteric viruses, *Cryptosporidium*, and *Giardia*, respectively in drinking water and an annual risk benchmark of 10^{-4} or 1 infection per 10,000 people per year (USEPA, 2017). Several studies on QMRA consistently predicted that PWSs under de facto reuse scenarios (containing $\geq$ 10% of treated wastewater effluent in a surface water source) exceeded the annual microbial human health risk benchmark (Amoueyan, Ahmad, Eisenberg, Pecson, & Gerrity, 2017; Chaudhry, Hamilton, Haas, & Nelson, 2017; Lim, Wu, & Jiang, 2017; Soller, Eftim, & Nappier, 2019). In contrast, direct potable reuse (DPR) scenarios were several orders of magnitude below the risk benchmark. A study by Chaudhry et al. (2017) estimated that blending even 1% of a surface water source containing 50% DFR with advanced treated effluents at a conventional PWS can significantly increase high annual risks.

The environmental retention time in the surface water is the amount of time between the wastewater outfall to surface water and drinking water intake (Reuse, 2012). QMRA studies by (Amoueyan et al. (2017); Lim et al. (2017)) indicated that the residence time of pathogens in an environmental buffer (e.g., lake, reservoir) can be a critical factor that reduces the annual risks. Soller et al. (2019) implied this role of the environmental retention time in surface water. Water quality is impacted by in-stream contaminant attenuation processes and environmental travel time between the locations where the treated effluent enters surface water and drinking water intakes (Reuse, 2012). Understanding de facto reuse in the extent of attenuation of contaminants and travel time in surface water can mitigate the public health risks associated with de facto (unplanned) potable reuse. The quantification of de facto reuse present in potable water sources across the U.S can enhance the understanding of epidemiological and risk assessment studies to support the implementation of direct potable reuse schemes (advanced treated reclaimed water) that may offer significant reductions in public health risks.

A geospatial model De facto Reuse Incidence in our Nations Consumptive Supply (DRINCS) was previously developed and validated to fulfill the top research need of the National Research Council associated with a systematic analysis of the extend of the nation’s de facto potable reuse (Rice & Westerhoff, 2015; Rice, Wutich, & Westerhoff, 2013). However, the DRINCS model only estimated the level of de facto potable reuse (DFR) for 2,056 surface water intakes at 1,210 larger DWTPs (serving > 10,000 people) in the United States. Results indicated that the high frequency of DFR was observed at more than 50% of drinking water intakes for these larger water systems but with a relatively low magnitude of less than 1% of treated municipal wastewater under mean annual streamflow condition. **Compared to larger DWTPs, there are nearly three times as many surface water intakes at smaller DWTPs serving ≤ 10,000 people as those at larger systems (N = 6,045 surface water intakes at 3,984 smaller DWTPs) (USEPA, 2019b)**. Yet very little data is available to estimate the occurrence of de facto reuse associated with smaller DWTPs. While the risks of those systems for CECs originate from treated wastewater in finished drinking water may be more significant than larger water systems. Thus, quantification of de facto reuse at smaller DWTPs can facilitate a full understanding of de facto reuse impacts on drinking water supplies at larger versus smaller DWTPs and their current capability of water treatment technologies can have the potential to mitigate the public health risks associated with exposure to CECs.

The overall goal of this dissertation is to quantify levels of de facto reuse at surface water intakes for smaller DWTPs (serving ≤ 10,000 people) across the United States by expanding the De facto Reuse Incidence in our Nations Consumptive Supply (DRINCS) model and develop a programmed ArcGIS model tool capable of proximity analysis between upstream WWTPs and DWTPs. **The hypothesis tested was whether smaller DWTPs (serving ≤ 10,000 people) in the United States are disproportionally dependent upon treated wastewater and lack advanced technology capable of**

removing CECs in source water compared to larger water systems.

This dissertation addresses the following research questions: What is the extend of de facto reuse at smaller DWTPs across the United States and how to develop a proximity model tool in the DRINCS model? To answer the research question, there is a four-step approach: (1) Expanding the DRINCS model with the inclusion of all smaller DWTPs across the U.S and updating with the latest WWTPs database, (2) Develop a proximity analysis tool to determine the distance and travel time of CECs in surface water between upstream WWTPs and DWTPs, (3) Compare de facto reuse at all smaller versus larger DWTPs across the U.S under varied streamflow conditions, (4) Investigate the current capabilities of the treatment process at DWTPs to reduce the health risk associated with exposure to CECs to the public population.

Dissertation Objectives

1) Demonstrate how predicted DFR by the DRINCS model can be confirmed with field observation of CECs occurrence at DWTPs

2) Groundtruth location of all surface water intakes at DWTPs in the continental United States

3) Quantify and compare the level of de facto reuse for smaller versus larger public water systems across the United States by expanding a previous version of the DRINCS model

4) Develop an automated proximity tool to determine the travel times between multiple WWTPs and downstream DWTP

5) Advance the model with an automation process (the DRINCS version 2.0)

Dissertation Organization

This dissertation is organized into seven chapters. Chapter 1 gives an overall introduction leading to Chapter 2, which provides a literature review on the global occurrence of de facto reuse, treatment technology, and potential health risks associated with CECs in drinking water for different communities. Chapter 3 demonstrates how predicted DFR by the DRINCs model can be confirmed with filed observation of CECs occurrence at DWTPs (published in a peer-reviewed journal). Chapter 4 describes the development of DRINCS version 2.0. Chapter 5 gives details of a pilot study to quantify DFR at all sized DWTPs in Texas (published in a peer-reviewed journal). Chapter 6 illustrates an expansion of the study in Texas to a nationwide assessment of DFR by utilizing an updated version of the previous DRINCS model (in preparation for submission to a peer-reviewed). Chapter 7 synthesizes the dissertation summary and recommends future research directions.

Table 1.1 Dissertation Organization (list by publication status of chapters and respective objectives)

Objective 1: *Demonstrate how predicted DFR by the DRINCS model can be confirmed with field observation of CECs occurrence at DWTPs* • Dissertation Chapter 3 Published: **Nguyen, T**., Westerhoff, P., Furlong, E.T., Kolpin, D.W., Batt, A.L., Mash, H.E., Schenck, K.M., Boone, J.S., Rice, J. and Glassmeyer, S.T., 2018. Modeled de facto reuse and contaminants of emerging concern in drinking water source waters. Journal-American Water Works Association, 110(4), pp. E2-E18
Objective 2: *Quantify and compare the DFR at smaller versus larger DWTPs in Texas and develop proximity algorithms to estimate travel times between upstream WWTPs and DWTP* • Dissertation Chapter 5 Published: **Nguyen, T.T.,** and Westerhoff, P.K., 2019. Drinking water vulnerability in less-populated communities in Texas to wastewater-derived contaminants. *npj Clean Water*, 2(1), pp.1-9.
Objective 3: *Quantify and compare the nationwide DFR for smaller versus larger public water systems across the United States by expanding the previous DRINCS model* • Dissertation Chapter 6 In preparation for peer-reviewed submission to *Environmental Health Journal*: **Nguyen, T**., Westerhoff, P., Acero, J. Unplanned Water Reuse Impacts on Drinking Water for Smaller-versus Larger Surface Water Systems across the United States.

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CHAPTER 2

LITERATURE REVIEW

De Facto Potable Reuse in the United States

De facto potable reuse or unplanned potable reuse is defined by the National Research Council as “a drinking water supply that contains a significant fraction of wastewater effluent, typically from upstream wastewater discharges, although the water supply has not been permitted as a water reuse project” (Reuse, 2012). Numerous wastewater treatment plants discharge treated effluents into waters of the U.S. In the United States, over 15,000 wastewater treatment plants (WWTPs) discharge more than 70% of treated wastewater (or 21.3 billion gallons per day) into surface water (USEPA, 2016). Notably, example rivers receiving treated wastewater effluents reported at regional scales include the South Platte River in Denver, Colorado; the Schuylkill River in Philadelphia, Pennsylvania; the Quinnipiac River in Connecticut; the Santa Ana River in southern California; the Ohio River in Cincinnati, Ohio and the Occoquan Watershed in Washington D.C (B. Chen, Nam, Westerhoff, Krasner, & Amy, 2009; B. Chen, Westerhoff, & Krasner, 2008; Metcalf et al., 2007).

Source surface water contains a portion of treated wastewater effluents from upstream WWTPs. Most global quantifications for the fraction of de facto reuse (or “dilution ratio”, “dilution factor”) in water supplies use Equation 1.

$$DFR = \frac{\sum Q_{WW,i}}{Q_{SW}} x100\% \qquad (1)$$

Where $Q_{WW,\,I}$: the upstream WWTP design discharge flow (cfs)

Q_{SW}: the streamflow at a surface water intake by a DWTP (cfs)

In 1980, the first attempt was made by the US EPA to estimate wastewater effluent contributions to water supplies in the U.S based on a scoping study (Swayne, Boone, Bauer, & Lee, 1980). The study showed the top 25 most effluent-impacted DWTPs have 2-6% DFR under average flow condition, and up to 50% under low flow (7Q10) condition (annual lowest 7-day average streamflow within every 10-year occurrence). A later survey from the U.S. EPA, in 1991, reported that 23% of total treated wastewater discharging into surface waters constituted more than 10% DFR of the source surface waters under average flow condition (NRC, 2012). Several case studies have been conducted to quantify the DFR at a watershed scale. For example, the Trinity River supplying surface water to DWTPs was reported to comprise approximately 50% of treated wastewater (Fono, Kolodziej, & Sedlak, 2006). A local-scale study by Brooks, Riley, and Taylor (2006) estimated that 60% of surface waters in the EPA Region 6 (i.e., Arkansas, Louisiana, New Mexico, Oklahoma, and Texas) were effluent-impacted source waters and comprised of at least 10% DFR under low flow (7Q10) conditions. There has been no systematic analysis to quantify the percentage of treated wastewater effluents in surface water sources in the United States between 1980 and 2008.

A Geographical Information System (GIS) approach developed by Rice, Wutich, and Westerhoff (2013) has updated the 1980 EPA analysis with the calculated de facto reuse levels at DWTPs. Over 30 years, results estimated an increase of 68% (from 4.9 to 8.2 billion gallons per day) in the volume of discharged wastewater effluents into surface water, and an increase in the DFR at 17 of the top 25 most effluent-impacted DWTPs with the average DFR increasing from 4.9% to 6.2% in 1980 and 2008, respectively. This study also identified hotspot surface water sources with the highest DFR levels in the

U.S. Such as the Passaic River in New Jersey, Neshaminy Creek in Pennsylvania, Schuylkill River in Pennsylvania, and Ray Hubbard Lake in Texas.

This GIS approach analysis was later extended to include more than 1,200 largest DWTPs (serving > 10,000 people) in the United States (Rice & Westerhoff, 2015). The method developed an ArcGIS model, De facto Reuse Incidence in our Nation's Consumable Supply (DRINCS) model to assess the spatial and temporal relationships between DWTPs and WWTPs. The DRINCS model took advantage of the existing hydrology of watershed, e.g., the U.S Geological Survey National Hydrography Dataset USGS NHDPlus, data on wastewater discharges permitted by National Pollutant Discharge Elimination Systems (NPDES) historical streamflow from the USGS stream gauging stations. Over 2,000 DWTP intake sites at those large water systems (serving > 10,000 people) and over 16,000 WWTPs discharged outfalls from the Clean Watersheds Needs Survey (CWNS 2008) are incorporated into the DRINCS model (Rice & Westerhoff, 2015). The model predicted a high frequency of DFR with more than 50% of drinking water intakes at those larger systems impacted by DFR. The magnitude of DFR was relatively low under average flow conditions as comprising of less than 1% of treated wastewater effluent and increased under low flow 7Q10 conditions during July and October (up to 90% treated wastewater at 15 of the 37 DWTP sites) (Figure 2.1).

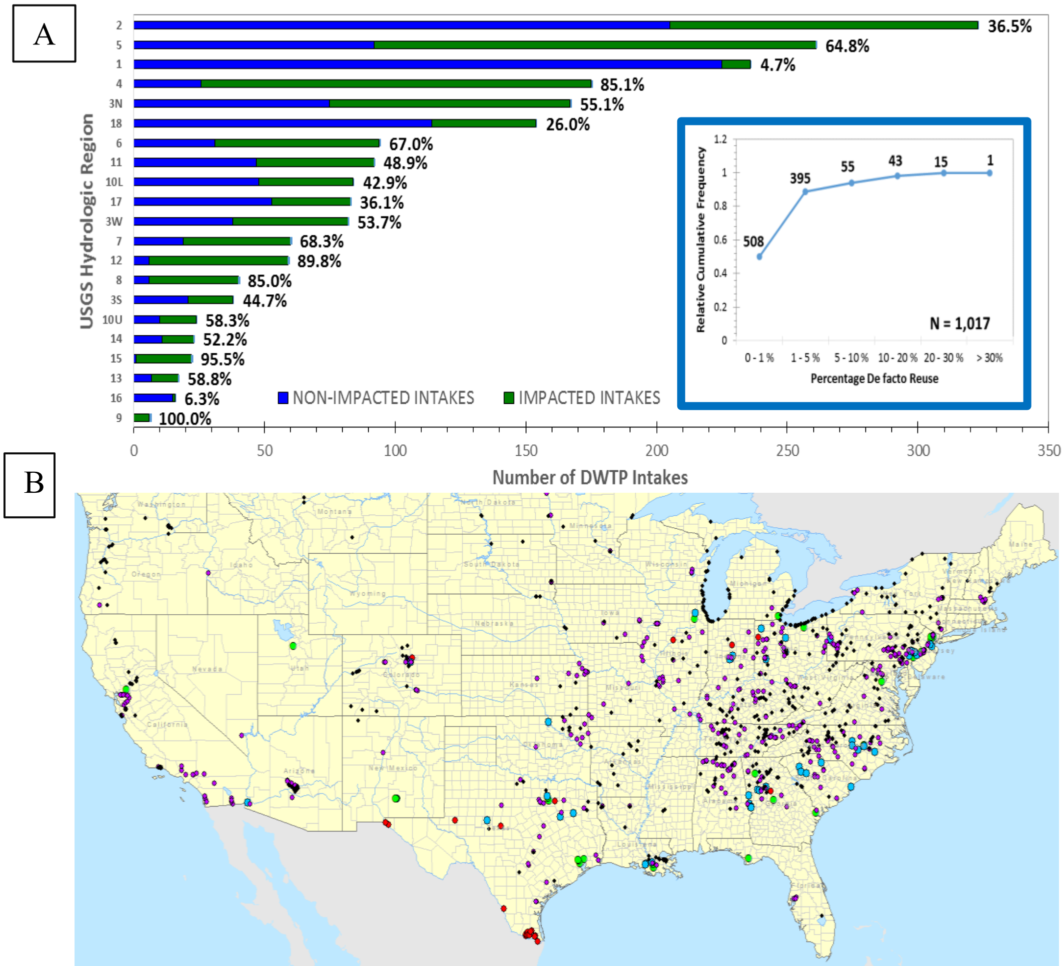

Figure 2.1 Distribution of De Facto Reuse at all Large DWTPs Serving more than 10,000 People in the United States

De Facto Potable Reuse Around the World

More than half of the world's population resides in urban areas in 2016 with about 4 billion people (UNDESA, 2016). This number is projected to increase to 60% with more than 5 billion people by 2030. This leads to an increase in the volume of municipal wastewater (raw sewage or treated at different levels) discharging into the surface waters. In comparison to developed countries, developing countries have lower sewerage coverage which collects and conveys industrial and municipal wastewater into treatment facilities and lack of centralized wastewater treatment plants (Mara, 2004). In the United States, approximately 75% volume of municipal wastewater has been collected into sanitary systems and more than half of the WWTPs employed secondary wastewater treatment treated (Reuse, 2012). Meanwhile, in Latin America, less than 15 percent of collected municipal wastewater from sewerage was reported to undergo any form of treatment before returning to water bodies by Mara (2004). The number is lower than the 10% of municipal wastewater collected in many developing countries in the Asia Pacific like Vietnam, Laos, and Cambodia (Marcotullio, 2007). Untreated domestic wastewater discharges directly to rivers or lakes, for example, the Saigon River in Ho Chi Minh city, Vietnam supplies 1.6 million m^3 per day (or 423 million gallons per days) for domestic water use (Minh, Phuoc, Quoc, Ngo, & Lan, 2016). In Syria, it was reported that untreated wastewater discharged directly from agricultural lands, leading to the degradation of surface water especially in the Barada River and Aleppo southern plains in Damascus (Melhem & Higano, 2011). The most polluted rivers in the world, which also supply drinking water in the world are located in Asia such as Bangladesh, the Ganges River/the Yamuna/Jamuna River in India, the Citarum River in Indonesia, and the Philippines (Kibria, 2015).

De facto potable reuse occurs globally as many communities currently use surface water sources containing wastewater discharged upstream. The most polluted river in Pakistan, the Ravi River, supplies about 11 million inhabitants with drinking water but also receives untreated municipal and industrial wastewater from five outfalls and two natural surface drains (Haider & Ali, 2010). In Bangladesh, the peripheral rivers surrounding Dhaka city which serve as drinking water sources for about 20 million inhabitants, is highly polluted due to municipal and industrial untreated effluents (Subramanian, 2004). More rivers and surface water bodies around the world are being impacted by upstream wastewater. For example, several rivers are in Japan (Simazaki et al., 2015), the Han River in Seoul South Korea (Yoon, Ryu, Oh, Choi, & Snyder, 2010), the Mankyung River in Jeollabuk-do of South Korea (Kim et al., 2009), and the Semenyih River in Putrajaya Malaysia (Praveena et al., 2018). In South Africa, de facto reuse occurs when Lake Rietvlei, a drinking water supply for the capital city of Pretoria, is comprised of secondary treated effluent from the Hartbeesfontein Sewage Purification Works (Oberholster, Botha, & Cloete, 2008). Several large rivers in Europe also experience the occurrence of de facto reuse. The River Thames, the second-longest river in the United Kingdom with a 215-mile length, was impacted by upstream wastewater from different point sources and non-point sources such as agricultural runoff, combined sewer overflows, and treated effluents from large WWTPs and industry (Blackburn, O'Neill, & Rangeley-Wilson, 2009). The Rhine River in central Europe supplying water for about 22 million people in six countries (Switzerland, Austria, Germany, France, Belgium, and the Netherlands), receives a sizable volume of treated effluents (Ruff, Mueller, Loos, & Singer, 2015).

The sixth-longest river in the world, the Murray River in Australia, supplies water for agricultural and municipal use for several regions of Victoria, New South Wales, and South Australia, contain a portion of treated wastewater effluents directly discharged from WWTPs upstream (Kumar et al., 2012). De facto reuse also occurs in cities along the Hawkesbury Nepean River in Australia (Khan & Anderson, 2018).

This unintentional scenario of de facto water reuse is likely widespread, but there is a lack of a systematic analysis of the extent of quantification of percent wastewater effluent in source surface water. Notably, a recent comprehensive study quantified the degree of wastewater impact in surface water supplying to agricultural irrigation in numerous watersheds in four EU countries (Spain, Italy, France, and Germany) (Drewes, Hübner, Zhiteneva, & Karakurt, 2017). Results indicated that two rivers from the Ebro River basin in the northeastern of Spain contained 3 % to 11% treated wastewater effluent under average flow condition; and the DFR level ranged between 8 and 82% under average flow condition in the Llobregat River Basin (Drewes et al., 2017). In Italy, the Adda River and Oglio River in the Po watershed are major surface water sources for agricultural irrigation which are comprised of 4 and 15% treated effluents, respectively under average flow conditions (Drewes et al., 2017). In France, the Loir River basin showed significant dilution with a low DFR range between 0.3 and 2.6% under average flow conditions, while the Montpellier River basin had a higher DFR range between 13% and 51% under average flow conditions (Drewes et al., 2017).

Surface water is also supplied for artificial groundwater recharge based on induced bank filtration and surface spreading methods. The same calculation of de facto reuse at surface water supplies (Equation 1) is used for these artificial groundwater recharge sites in two study cases in Toulouse, France, and the City of Berlin, Germany (Drewes et al., 2017). The findings concluded that the managed aquifer recharge (MAR) sites along the Garonne River and Ariege river in France were impacted by a low DFR under average flow conditions with a range of 0.4 to 1.4% and 0.41%, respectively. A higher level of DFR with more than 20% of treated wastewater under average flow conditions was predicted at these MAR sites around lake Tegel, Germany to provide drinking water for 3.7 million people in the city of Berlin (Pekdeger et al., 2006).

Droughts or low flow conditions of rivers can lead to an increase in the percentage of wastewater contributions to overall river flows and increase impacts on potable water quality. Several studies have attempted to quantify the de facto reuse in drinking water supplies at a local watershed. A hydrological model effort predicted the percentage of effluents from sewage treatment works to receiving rivers in the River Ouse watershed, United Kingdom. Results showed that most rivers used as drinking water supplies in this studied basin had more than 25% of treated wastewater effluent under dry weather flow (Q90 low flow) conditions in the Cambridge area, United Kingdom (Reuse, 2012). A study in Spain estimated the dilution factor at surface water intakes along the Llobregat river with a DFR range from 12% to 25% in wet and dry years, respectively. These surface water intakes supply water for approximately 1.8 million people in Catalonia, northeast Spain (Mujeriego, Gullón, & Lobato, 2017).

In Asian countries, even where there is a limited dataset on infrastructure and frequent lack of long term historic streamflow, research by Wang, Shao, and Westerhoff (2017) showed the efforts to estimate DFR for the river reaches in the Yangtze River in China which provides drinking water supplies for over a 1/15th of the world’s population. Results estimated an increase in the volume of wastewater effluent from 8% to 14% between 1998 and 2014 under low flow (7Q10) conditions. The highest DFR can be observed at the outlet of the watershed (Shanghai city) because of the highest accumulation of wastewater, and at the central area of the Yangtze River basin due to the high population and lower stream flows.

To the authors' knowledge, internationally, there have been only three nationwide assessments of quantification of de facto reuse in surface water supplies. In Switzerland, a nationwide assessment estimated DFR under dry weather flow (Q347) conditions. In Switzerland, Q347 is defined in Article 4 of the Water Protection Law as "the rate of flow which, averaged over ten years, is reached or exceeded on an average of 347 days per year and which is not substantially affected by damming, withdrawal or supply of water " (http://hydrologicalatlas.ch). Q347 is used to calculate the environmental flow to establish three protection levels of streamflow in Switzerland. Q347 flow rate corresponds to the quantity of water which is reached or exceeded in 95% of cases (Q95). The low flow Q95 is chosen and widely used in Europe as it is relevant to various topics in water resources management. Figure 2.2 shows the de facto reuse at surface water supplies under Q347 flow (Drewes et al., 2017; Karakurt, Schmid, Hübner, & Drewes, 2019).

Figure 2.2 Geographical Distribution of Treated Wastewater in Streams under Dry Weather Low Condition (Q347) in Switzerland

In a national reconnaissance study in Germany, river reaches were estimated to constitute more than 30-50% treated wastewater effluents under mean minimum discharge conditions between May and September of the year (Karakurt et al., 2019). A nationwide survey on Japanese rivers has reported the Freshness Level (shown in Figure 2.3) of receiving rivers which are calculated as the ratios of natural or pristine surface water flow (without wastewater) to total river flow at DWTP intakes. In figure 2.3, the blue color indicates 90-100% or less than 10% de facto reuse; green color indicates 50 – 90% or less than 50% de facto reuse; yellow color indicates less than 50% or more than 50% de facto reuse. This calculation of Freshness Level is equaled to 100% minus DFR at a drinking water intake. The estimation for two years, 2003 and 2004 showed a higher Freshness Level of rivers with a range of 27% and 79%, or a DFR range between 21% and 73% (MLIT, 2019).

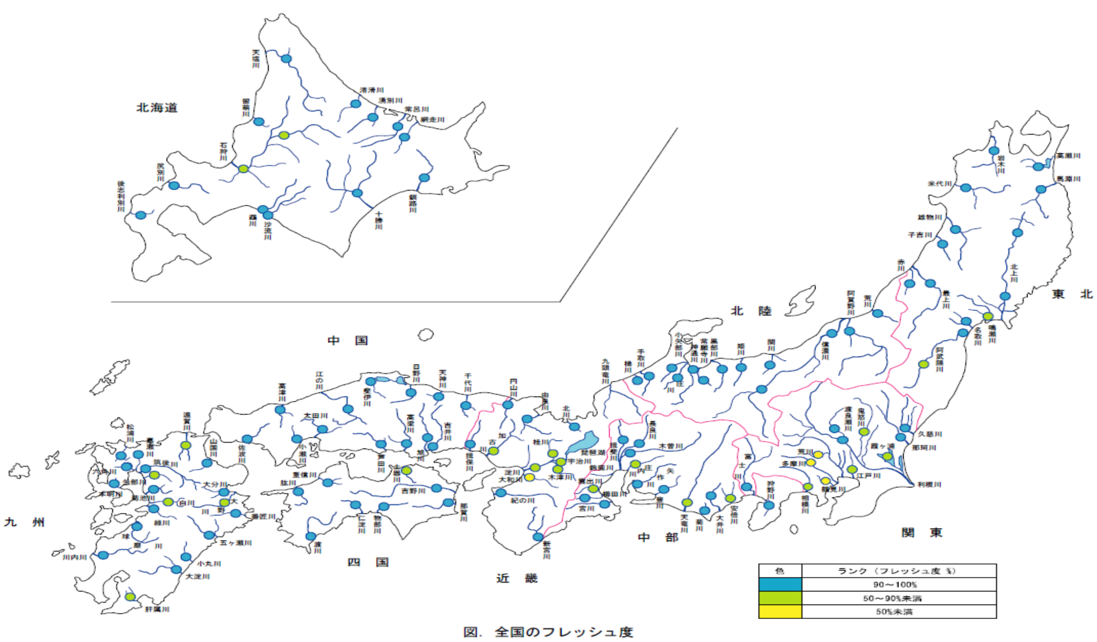

Figure 2.3 A Nationwide Survey of Freshness Level at Drinking Water Treatment Plants in Japan

Technology Employed in Planned Potable Reuse

The wide practice of de facto reuse reflects the scenarios of wastewater is likely to return to the water supply. The reuse of treated municipal wastewater (reclaimed water) as an alternative potable water supply has significant potential for helping freshwater shortage in some areas and reducing the human health risks by de facto reuse in drinking water.

Two categories in planned potable reuse are indirect potable reuse (IPR) and direct potable reuse (DPR) (Reuse, 2012). In both schemes, secondary treated effluents first undergo advanced engineered treatment to produce highly treated reclaimed water. However, compared to DPR, IPR includes an additional environmental buffer (e.g., lake, reservoir, and groundwater aquifer) to store and blend highly treated reclaimed water with the primary drinking water source before being treated at a conventional water treatment system (Leverenz, Tchobanoglous, & Asano, 2011). Direct potable reuse (DPR) relies upon an advanced wastewater treatment plant to produce reliable and highly treated reclaimed water suitable for direct human consumption.

Over the past decade, the development of advanced engineered treatment results in the ability to remove multiple contaminants including nutrients, organic matter, total dissolved solids (TDS) or salinity, pathogens, CECs, and pathogens in conventional (secondary) treated wastewater (Reuse, 2012). Nutrients can be reduced by several processes including biological nitrification and denitrification, gas stripping, breakpoint chlorination, and chemical precipitation. Organic matter can be removed by various advanced processes including activated carbon, chemical oxidation (using ozone, advanced oxidation process AOPs), nanofiltration NF, and reverse osmosis RO (Reuse, 2012). Softening, electrodialysis, NF, and RO are capable of removing total dissolved solids (TDS) in secondary treated wastewater. RO and NF are classified as low-pressure RO products, using pressure-driven membrane separation processes to remove dissolved solutes, i.e. ions and molecules such as sodium, chloride, calcium, magnesium, dissolved natural organic matter NOM, and synthetic organic chemicals from a solution (Reuse, 2012). AOPs include ozonation and/or combined with hydrogen peroxide, and UV light and/or combined with hydrogen peroxide (UV/H_2O_2) to produce hydroxyl radicals, then effectively oxidize CECs (Westerhoff, Yoon, Snyder, & Wert, 2005). Various processes can be combined to achieve the target removal depending on source water quality, reuse requirements, etc. (Reuse, 2012).

In the U.S, the use of reclaimed water for drinking water supplies (potable reuse schemes) has been implemented since 1962 when the first project of groundwater recharge via soil-aquifer treatment in Montebello Forebay in County Sanitation Districts of Los Angeles County in California (Reuse, 2012). Other IPR pioneering projects are groundwater recharge via seawater barrier in Water Factory 21, Orange County California (1976), and surface water augmentation in the Upper Occoquan Service Authority (1978) (Reuse, 2012). In 2010, approximately 355 MGD (1,350 m^3/d) of reclaimed water was reused for planned potable reuse schemes in the U.S. which accounts for about 0.1% of the volume of municipal wastewater currently being treated (Reuse, 2012). Currently, there have been a total of 49 planned potable reuse projects which are in operation or under study (shown in Figure 2.4), and most of these IPR projects are located in arid southwest of the U.S (water-scarcity regions), such as California, Texas, Arizona and Florida (Reuse, 2012).

Ion Exchange; LC – Lime Clarification; MBR – Membrane Bioreactor; MF - Microfiltration; O3 – Ozone Disinfection; PAC – Powdered Activated Carbon; RO – Reverse Osmosis; UF - Ultrafiltration; UV – Ultraviolet Radiation

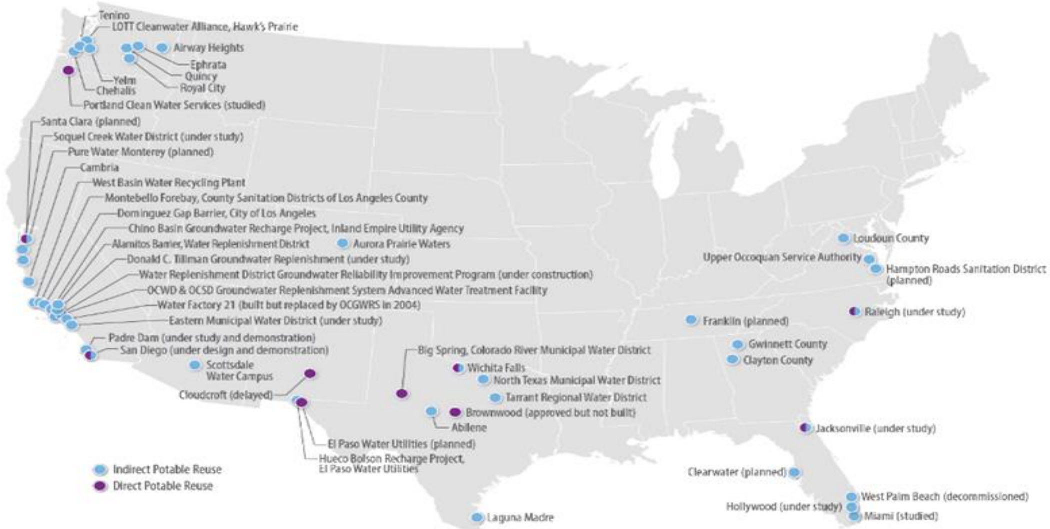

Figure 2.4 Current Status of Potable Reuse Projects in the United States

All over the world, it is reported to have 25 potable reuse projects (Figure 2.5) and most of them are located at high water stress areas such as Australia, the Middle East, and the Mediterranean (Reuse, 2012).

Figure 2.5 Examples of IPR And DPR Projects in the World (excluding the United States)

Australia. The first full-scale groundwater replenishment scheme in the northern suburbs of the Western Australian capital Perth is operated by Western Australia's Water Corporation. Highly purified effluents by ultrafiltration, reverse osmosis, and UV-disinfection discharge into managed aquifer recharge and the construction commenced in 2014 (Khan & Anderson, 2018). Another constructed IPR scheme (but now is being decommissioned) in Australia is the Western Corridor Recycled Water Scheme in South East Queensland. The advanced wastewater treatment includes microfiltration (MF) and reverses osmosis RO followed by an advanced oxidation system (AOP) using UV light and hydrogen peroxide (Khan & Anderson, 2018). The Saint Mary Advanced Water Recycling Plant in New South Wales, Australia, with the full-scale operation in 2010, treats municipal wastewater by ultrafiltration and reverse osmosis which then is discharged to supplement the flows of the Hawkesbury-Nepean River (17 km upstream of the town of Richmond) (Khan & Anderson, 2018). Other potable reuse schemes in Australia, however, are not successful, are the project in the City of Toowoomba, Queensland failed to construct, and Goulburn, New South Wales, the Australian Capital Territory did not proceed (Burgess, Meeker, Minton, & O'Donohue, 2015).

South Africa. The City of Windhoek in Namibia has constructed the world's first potable water treatment plant named Goreangab Water Reclamation Plant (WRT) in 1969 which serves approximately 250,000 people (Z. Chen, Ngo, & Guo, 2013; Metcalf et al., 2007). In 2002, a new Goreangab WRT used advanced technologies such as ozonation and membrane filtration was constructed to provide 35% of the daily potable reuse for the city (Z. Chen et al., 2013). There have been more planned reuse projects recently implemented in South Africa (Burgess et al., 2015). For example, the city of George with a 10ML per day (or 2.64 MGD) IPR plant using ultra-filtration and powdered activated carbon in 2009; the town of Beaufort West with a 2.3 ML per day (or 0.61 MGD) DPR plant using ultrafiltration, two-stage RO, permeate disinfected by ultraviolet light (UV) in 2010; the City of Cape Town with a 20ML per day (or 5.28 MGD) membrane bioreactor (under construction); Port Elizabeth with a 45 ML per day (or 11.89 MGD) membrane bioreactor for industrial and/or indirect potable reuse (under construction); the town of Hermanus with a 5ML per day (or 1.32 MGD) DPR plant using ultrafiltration, RO, advanced oxidation and carbon filtration (Burgess et al., 2015).

Singapore. The Singapore Water Reclamation Study was constructed in 2000 and until now, a total of five NEWater (advanced treated wastewater in Singapore) operational plants located at Bedok, Kranji, Seletar, Ulu Pandan, and Changi can provide 15% of water demand supplying directly to industries (Z. Chen et al., 2013). About 6% of the NEWater blended with raw water in reservoirs and supplied 1% of the total potable water supply. By 2020, the NEWater will continue to expand the capacity to 284 ML/day (or 75 MGD) which accounts for 40% of the total water supply (Z. Chen et al., 2013; Metcalf & Eddy et al., 2007).

Europe. The Langford Recycling Scheme in Essex and Suffolk in the United Kingdom was the first project operated in 1997 using microfiltration and ultraviolet in a tertiary WWTP. The treated effluent discharged into the Chelmer river was used for flow augmentation and drinking water supplies for 100,000 population (Z. Chen et al., 2013). The Torreele IPR project in Wulpen Belgium was constructed in 2002. The advanced processes include microfiltration, reverse osmosis, and ultraviolet which can produce 40-50% of potable supply serving more than 60,000 people (Rodriguez et al., 2009).

Wastewater Treatment

Effluent Permit Program. In the United States, the 1972 Federal Water Pollution Control Act (Clean Water Act) established the National Pollutant Discharge Elimination System (NPDES) permit program to achieve the goal of: "water quality [that] provides for the protection and propagation of fish, shellfish, and wildlife and provides for recreation in and on the water" (USEPA, 1972). Under the Title 40 of the Code of Federal Regulations (CFR) at Part 122, the U.S. Environmental Protection Agency (USEPA) administered the NPDES program to regulate point source discharges into the nation's waters such as publicly owned treatment works (POTWs). The US EPA (in 40 CFR part 133) set the secondary treatment as the national standard for permitted municipal wastewater effluents discharging into surface water.

Technology-based limitations and water quality-based limitations are two types of control in the NPDES permit (USEPA, 2010). The required standards include five-day biochemical oxygen demand (BOD_5), total suspended solids (TSS) removal, and pH and levels of treatment for each effluent discharge. For example, the NDPES permit regulates municipal effluents based on a 5-day biochemical oxygen demand (BOD_5) concentration (30-day average) with greater than 45 mg/L (primary treatment); greater than 30 mg/L but less than or equal to 45 mg/L (advanced primary); less than or equal to 30 mg/L and total suspended solids and pH (secondary treatment); less than or equal to 20 mg/L (advanced treatment) (USEPA, 2010).

Publicly Owned Treatment Works. The U.S. EPA in compliance with the Clean Water Act (CWA) section 516(b)(1)(B) has prepared the Clean Watersheds Needs Survey (CWNS) Report to Congress to estimates the capital investment necessary to ensure POTWs meet the CWA regulations (USEPA, 2016). Nearly 15,000 WWTPs serve about 76% percent of the U.S. population (with approximately 238.2 million people) in 2012 (USEPA, 2016). The reported nation's total wastewater flow from POTWs was 46,872 MGD and 62.75% of treated effluents (or 29,410 MGD) were discharged into inland waterways (USEPA, 2016). About 1.3 percent of POTW effluents employed less than secondary treatment before discharging into surface water by USEPA (2012) while more than half of POTWs employed conventional secondary treatment (USEPA, 2016). Primary and secondary wastewater treatment processes are not capable of removing organic matter, contaminants of emerging concern, and pathogens in raw wastewater (Metcalf et al., 2007).

Wastewater Treatment Process. At a publicly owned treatment work, municipal wastewater typically undergoes preliminary treatment, primary treatment, secondary treatment (without or with nutrient removal) (Tchobanoglous, Burton, & Stensel, 2003). Tertiary/or advanced treatment may be required at specific WWTPs (Figure 2.6).

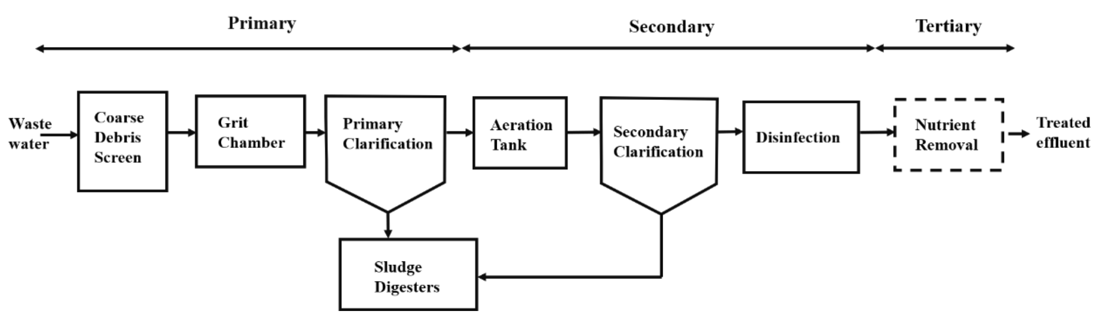

Figure 2.6 Diagram of Wastewater Treatment Processes in a Wastewater Treatment Plant

Preliminary wastewater treatment includes influent flow measurement, screening, and grit removal to separate heavy, inorganic, sand-like solids, coarse solids such as rags from municipal influents (Reuse, 2012). Primary wastewater treatment comprises of primary settling, and gravity sedimentation to remove floating debris and solids in municipal wastewater. The primary treatment process is capable of removing more than 50% of the suspended solids, 30% of total biochemical oxygen demand (BOD), and some effective reduction in nutrients, pathogenic organisms, trace elements, and potentially toxic organic compounds (Reuse, 2012).

Secondary wastewater treatment consists of chemical and biological processes (such as aerated activated sludge basins, or fixed-media filters (e.g., trickling filters, rotating bio-contactors) to remove total suspended solids, dissolved organic matter (BOD), and nutrients (Tchobanoglous et al., 2003). Not all secondary treatment is followed by tertiary or advanced wastewater treatment. Tertiary or advanced wastewater treatment is employed only when higher quality treated effluent (than secondary treated standards) is required to protect the receiving water conditions or other uses.

Tertiary wastewater treatment is capable of removing nutrients (nitrogen, phosphorus), ammonia, organic matter (BOD), CECs, total dissolved solids (TDS), salinity, and pathogens in secondary treated effluents (Reuse, 2012). In tertiary wastewater processes, the nitrification process can remove ammonia, and nitrification followed by denitrification can remove nitrogen. Phosphorus can be removed by microbial uptake or chemical precipitation process. Filtration adds coagulants to reduce TDS and associated BOD in the secondary-treated effluent. Activated carbon adsorption can remove organic matter and CECs while disinfection can inactivate/remove pathogens. The use of combined treatment unit processes at the primary and secondary level can achieve the same efficiency as advanced wastewater treatment, for example, in nutrient removal. There are many variations in employing these levels of wastewater treatment. For instance, primary treatment is eliminated in some treatment train and long-term retention in lagoons is sometimes an alternative for both primary and secondary treatment (Reuse, 2012).

Drinking Water Treatment

Public Water System. In the U.S, public water supply from a surface water source (river, lake, or reservoir) accounts for more than 60% of the total use of surface water with 39,000 Mgal per day (Dieter, 2018). The Safe Drinking Water Act (SDWA) was created by Congress in 1974 to protect the nation's public water supply sources and ensure safe and clean water quality for human consumption (USEPA, 1996b). Under the authority of SDWA, the US EPA established the National Primary Drinking Water Regulations which set standards on maximum contaminant levels for contaminants in drinking water, available water treatment technology and the requirement for monitoring and sampling contaminants.

The Safe Drinking Water Information System (EPA SDWIS) is a federal database warehouse created under the 1996 SDWA amendment and the US EPA to maintain the drinking water information periodically reported by all states (USEPA, 1996a). The SDWIS contains basic water facility inventory, violations, and enforcement for each public water system. Public water systems are characterized by ownership type (public or private), type of source water (surface water, groundwater, and purchased water), and population served (USEPA, 2019c). Public ownership includes government (federal, state, or local) and Native American tribes. Public water systems are classified as community water systems (CWS), transient non-community (TNC), and non-transient non-community (NTNC) based on whether they serve the same customers year-round or on an occasional basis. A public water system may have several types of source water, but if the system has any surface water source, it is classified as a surface water system. A purchased water system (groundwater purchased or surface water purchased) obtain treated water from other public water systems, and most commonly, from surface sources. PWS size is categorized based on the number of the population served for each PWS. This is the local retail population served and does not include the wholesale population for purchasing water from another PWS (USEPA, 2019c). Following the US EPA, water systems are categorized into five sizes based on the number of the population served: very small systems serve < 500 people, small serve 501-3,300, medium serve 3,301-10,000, large serve 10,001-100,000 and very large serve > 100,000 people (USEPA, 2019c).

In the United States, a total of 149,057 public water systems (PWSs) are serving about 90% of the U.S. population (or more than 300 million people) (USEPA, 2019c). Table 2.1 shows the number of public water systems categorized by type of water system and type of source water in the calendar year 2018. One-third of all the public water systems are community water systems serving the same year-round population (N = 50.094) and two-thirds are non-community water systems (N = 98,963), including transient systems (N = 81,023) and non-transient systems (N = 17,940).

Table 2.1 Public Water Systems in the U.S Categorized by Type and Number of Systems in the Calendar Year 2018

Type of Public Water Systems	**Number of active systems**
Community Water System	50,094
Non-Transient Non-Community Water System	17,940
Transient Non-Community Water System	81,023
Total	**149,057**

More than 90% of the U.S population (312 million people) is supplied drinking water from a community water system (Table 2.2) (USEPA, 2019c). Compared to the groundwater source, the number of CWSs using surface water is one-third smaller (N = 11,800), but surface water CWSs serve a higher population (66 % of the U.S population).

Table 2.2 Public Water Systems Categorized by Size or Number of Populations Served and Type of Water Source in the Calendar Year 2018

Community Water System		**Type of water source**	
Size	**Population served**	**Surface water**	**Groundwater**
Very Small	< 500	3,155	24,196
Small	501 – 3,300	3,666	9,741
Medium	3,301-10,000	2,216	2,774
Large	10,001-100,000	2,398	1,510
Very Large	> 100,000	365	73
Total Number	50,094	11,800	38,294
Total Population Served	312,590,152	221,499,433	91,090,719

Drinking Water Standards. The 1976 Safe Drinking Water Act, under Title 40 of Code of Federal Regulations CFR Part 141, created the framework to establish the

National Interim Primary Drinking Water Regulations, National Primary Drinking Water Regulations (legally enforceable standards), and National Secondary Drinking Water Regulations (non-enforceable guidelines) to protect drinking water sources and ensure clean and safe water supply to the U.S population (USEPA, 1996a). The US EPA set national regulations (or rules) and primary standards for public water systems to limit levels of more than 90 contaminants in drinking water to protect human consumption (USEPA, 1996a). The six main groups of these regulated contaminants in drinking water include microorganisms, disinfectants, disinfection byproducts, inorganic chemicals, organic chemicals, and radionuclides.

Pathogens, such as *Giardia*, *Cryptosporidium*, and viruses, are often found in source water and can cause gastrointestinal illness (diarrhea, vomiting, cramps, and other health risks). The Surface Water Treatment Rules (SWTR) is applied to all public water systems using surface water to reduce illnesses caused by pathogens in drinking water. The SWTRs requires water systems to filter and disinfect surface water sources to achieve 2-log removal/inactivation (99%) of *Cryptosporidium*, 3-log removal/inactivation (99.9%) of *Giardia lamblia*, and 4-log removal/inactivation of viruses (99.99%). Some rules within the suit of SWTR are specific to different sizes of public water systems using surface water. The 2002 Long Term 1 Enhanced Surface Water Treatment Rule (LT1ESWTR) controls *Cryptosporidium* with 2-log removal (99%) at only large surface water PWSs serving more than 10,000 people (USEPA, 2002). More recently, the 2006 Long Term 2 Enhanced Surface Water Treatment Rule (LT2ESWTR) addresses the health effects associated with *Cryptosporidium* in surface water supply and requires all sizes of surface water public water systems to monitor and sample *Cryptosporidium* and employ additional treatment at higher risk systems based on sampling (USEPA, 2006). Some states require treatment of 10 log oocyst *Cryptosporidium* and 10-log *Giardia* cyst removal in IPRs using recycled water via spreading basins or direct injection of surface water to groundwater, such as California and Nevada (10 log-removal of *Cryptosporidium*) (USEPA, 2017). These log removals are intended to achieve a risk benchmark of 1 infection per 10,000 people per year for *Giardia lamblia* and *Cryptosporidium* spp. presence in raw sewage (Macler, A, & Regli, 1993; Metcalf et al., 2007; Sinclair, O'Toole, Gibney, & Leder, 2015).

Disinfection byproducts and disinfectants are being regulated under the Comprehensive Disinfectants and Disinfection Byproducts (DDBR) Rules (Stage 1 DDBR and Stage 2 DDBR) to reduce drinking water exposure to disinfection byproducts from the public population (USEPA, 1998). Some disinfectants and disinfection byproducts (DBPs) have been shown to cause cancer and reproductive effects in lab animals and suggested bladder cancer and reproductive effects in humans. Under comprehensive DDBR rules (Stage 1 and Stage 2), regulated DBPs are trihalomethanes (THM), haloacetic acids (HAA), chlorite, and bromate. The regulated maximum concentration level (MCL) in drinking water for these regulated DBPs is 0.08 mg/L, 0.06 mg/l, 1 mg/L and 0.010 mg/L, respectively (USEPA, 2019a). This rule is applied to all community water systems that use disinfectants other than UV light. The regulated disinfectants under this rule are chlorine, chloramines, and chlorine dioxide with the maximum residual disinfectant level (MRDL) at 4 mg/L as Cl_2 for both chlorine and chloramines, and 0.8 mg/L for chlorine dioxide (USEPA, 2019a).

Over 65 chemical contaminants are regulated under the 1989 Phase II/V Rules or the Chemical Contaminant Rules including three groups: inorganic contaminants (IOCs) (including arsenic and nitrate), volatile organic contaminants (VOCs), synthetic organic contaminants (SOCs) (USEPA, 2019a). This rule applies to all public water systems and the MCL is set for each contaminant. Radionuclides are regulated under the 2000 Radionuclides Rule to reduce drinking water exposure to radionuclides. Radionuclides in drinking water can cause the risk of cancer, and toxic kidney effects of uranium. Radionuclides include combined radium-226 and radium-228, gross alpha particle radioactivity, beta particle, and photon activity, and uranium. The regulated MCLs is 5 pCi/L, 15 pCi/L, 4 mrem/yr, 30 µg/L, respectively (USEPA, 2019a).

The US EPA established the Unregulated Contaminant Monitoring Rule (UCMR) to collect data for contaminants that may present in drinking water and do not have health-based standards set under the SDWA (USEPA, 2019b). Every five years, the US EPA makes a report on a Contaminant Candidate List (CCL) of 30 or fewer unregulated contaminants to be monitored nationally. These contaminants in the CCL list are not being regulated yet and not subject to any drinking water standards but maybe aware of their presence in public water systems for future regulation under the SDWA (USEPA, 2019b).

The fourth Unregulated Contaminant Monitoring Rule (UCMR4) was finalized in 2016 and currently provides national monitoring data of 30 contaminants. The Fourth Contaminant Candidate List-4 (CCL-4) contains 97 chemicals or chemical groups and 12 microbial contaminants (USEPA, 2019b). EPA is currently working on the UCMR-5 and plans to publish the final rule by December 2021. The most recent notice was a proposed rule for perchlorate and exposure to perchlorate may be linked with inadequate iodine consumption, impairing the thyroid's ability and the potential effect on brain development in humans (USEPA, 2020). Perchlorate was originally on the CCL-1, 2, and 3 lists and UCMR-1. However, no maximum contaminant level (MCL) has been set since the EPA report in 2001, and perchlorate is still not yet regulated (USEPA, 2020).

Surface Water Treatment Process. A surface water treatment plant employs a sequence of water treatment processes to remove contaminants from surface water sources (Figure 2.7) (Howe, Hand, Crittenden, Trussell, & Tchobanoglous, 2012). A typical conventional surface water treatment plant in the USA consists of screens, coagulation and flocculation, sedimentation, granular filtration, and disinfection process.

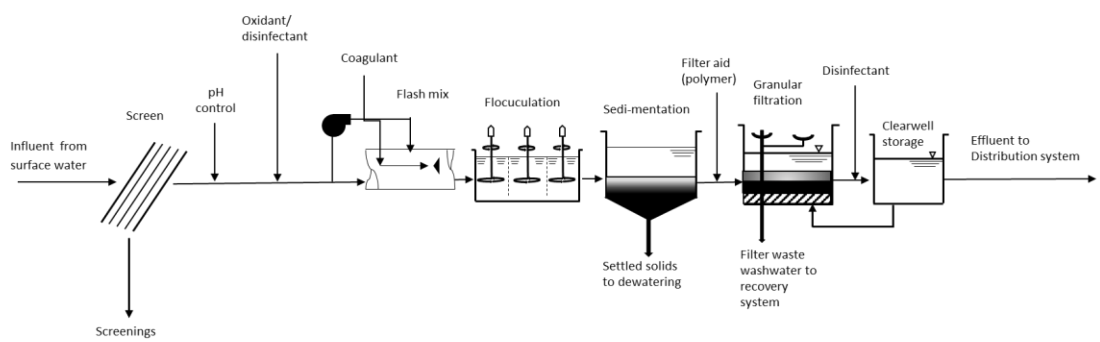

Figure 2.7 Typical Treatment Unit Processes for a Surface Water Treatment Plant

In the pretreatment process, large solids, debris, rags, or fish in raw surface water are removed by using coarse or traveling screens (Howe et al., 2012). Other pretreatment processes are using powdered activated carbon (PAC) for taste and odor control and synthetic organic carbon (SOC) removal, adding oxidants for iron and manganese removal, and using lime softening for hardness removal. Particulate matter and a portion of dissolved natural organic matter (NOM) in surface water can be removed by a coagulation and flocculation process, followed by sedimentation and/or filtration (Howe et al., 2012). Commonly used coagulants are aluminum sulfate (alum), ferric chloride, or ferric sulfate to destabilize suspended and colloidal particulate matter during flocculation. Larger flocs are subsequently settled out by gravity sedimentation and or/filtration (Howe et al., 2012). Filtration is widely used in DWTPs for the removal of solid particles (algae, sediment, clay, other organic and inorganic particles, etc.) through a porous medium. The most common filtration process is granular filtration using sand (rapid or slow filtration) (Howe et al., 2012). In deep filtration, smaller particles can be captured in the pores of the bed. The more recent technology is membrane filtration including microfiltration (MF) or ultrafiltration (UF) membranes (Howe et al., 2012).

Oxidation is an important process to disinfect pathogens in surface water and reduce chemical pollutants. The most common oxidants in water are chlorine, chlorine dioxide, and ozone which can destroy the cellular protein, nucleic acid, and cell wall or membrane of microorganism (Yoo, 2018). NOM and some inorganic constituents (e.g., perchlorate, arsenic, and some heavy metals) can be reduced by transferring from water to the surface of a solid in an adsorption or ion exchange process. Inorganic constituents include hardness (calcium and magnesium), nitrate, iron, and manganese can be effectively removed by an ion exchange process using synthetic resins (Howe et al., 2012). Granular or powdered activated carbon (GAC or PAC, respectively) are common used in an adsorption process to remove synthetic organic chemicals, taste- and odor-causing organics, color-forming organics, and disinfection by-product (DBP) precursors in surface water. The product water after undergoing surface water treatment processes is stored in a clear well before entering the distribution system (Howe et al., 2012)

Contaminant of Emerging Concerns. Several studies indicated that secondary treatment, or "standard technology" to treat municipal wastewater, is insufficient to eliminate contaminants in raw wastewater (Metcalf et al., 2007). Several contaminants present in secondary treated effluents such as organic precursors, disinfection by-products, contaminants of emerging concerns, and pathogens despite their low concentration (microgram per liter µg/L, nanogram per liter ng/L, or even picogram per liter pg/L) (Luo et al., 2014; Schwarzenbach et al., 2006). For several decades, numerous studies have focused on regulated contaminants in treated wastewater including polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), solvents, and pesticides (Belgiorno et al., 2007; Peakall, 1972). For the last two decades, the development of detection and analytical equipment has facilitated numerous studies on unregulated contaminants including pharmaceutical residues, endocrine disrupting compounds EDCs, personal care products, perfluorinated compounds, and plasticizers, referred to as chemicals of emerging concern (CECs) (Metcalf et al., 2007).

Disinfection by-products can be formed during the disinfection process in the presence of natural organic matter in secondary or tertiary wastewater treatment (Metcalf et al., 2007). The disinfection process includes chlorination, chloramination, ozonation, and ultraviolet radiation. The most common disinfection process is chlorination which adds dissolved chlorine gas or hypochlorite (i.e., bleach) to disinfect wastewater effluents or source water to a DWTP. Municipal wastewater is rich in nitrogen, iodine, and bromide which can promote the form of nitrogenous, iodinated, and brominated DBPs in treated effluents (Metcalf et al., 2007). For example, the formation of nitrogenous, iodinated, and brominated DBPs in effluents treated with ozonation by Huang, Fang, and

Wang (2005) or halogenated DBPs (e.g., trihalomethanes and haloacetic acids) from chlorination in treated effluents (Trussell, 1978). Chloramination disinfection process can promote the formation of non-halogenated DBPs such as N-nitrosamine in treated effluents (Krasner, 2009; Mitch et al., 2003); or in drinking water in compliance with the Stage 2 Disinfectants and Disinfection By-Products Rule (Seidel, McGuire, Summers, & Via, 2005). Ozone pretreatment is utilized to inactive pathogens and remove organic contaminants but also forms N-nitrosamine in drinking water during ozonation (Krasner, Mitch, McCurry, Hanigan, & Westerhoff, 2013).

Table 2.3 provides characteristics of seven CECs were selected as surrogates in the treated effluent (T. T. Nguyen & Westerhoff, 2019). Their natural attenuation in surface water was evaluated based on the DRINCS model results (further described in Chapter 5). Results indicated that with the distances between DWTPs downstream from multiple WWTP discharges ranging from <16 to >800 km, some CECs would be largely removed by natural attenuation while concentrations of more refractory CECs (e.g., TCEP, TCPP) remained unchanged at the downstream DWTP intakes (T. T. Nguyen & Westerhoff, 2019).

Table 2.3 The Concentration of Seven Contaminants of Emerging Concerns, Commonly Used as WWTP Surrogates, Detected in U.S Stream Samples

Compound	CAS number	Description	Median, ng/L	Maximum, ng/L
Tris(1,3-dichloro-2-propyl) phosphate (TCPP)	13674-84-5	flame retardant	180[2]	720[2]
Tris(2-chloroethyl) phosphate (TCEP)	115-96-8	flame retardant	100[1], 120[2], 280[3]	530[2]
Diclofenac	15307-79-6	nonsteroidal anti-inflammatory	1.1[2]	1.2[2]
Meprobamate	57-53-4	antianxiety	8.2[2], 242[3]	73[2]
Ibuprofen	15687-271	pain reliever	200[1], 49[3]	1000
Gemfibrozil	25812-300	anti-cholesterol	48[1], 2.2[2], 308[3]	790[1], 24[2]
Sulfamethoxazole	723-46-6	antibiotic	150[1], 12[2], 426[3]	1900[1], 110[2]

(1) (Kolpin et al., 2002)
(2) (Benotti et al., 2009)
(3) (Dickenson, Snyder, Sedlak, & Drewes, 2011)

Municipal wastewater contains a wide variety of pathogenic organisms which may be excreted by human and animal with infectious disease (Metcalf et al., 2007). Genus *Salmonella* is the most common bacterial pathogens in wastewater and protozoa *Cryptosporidium* parvum and *Giardia* lamblia are most resistant to conventional chlorination disinfection process, especially for the oocysts of *Cryptosporidium* parvum and the cysts of *Giardia* lamblia which are highly detected in wastewater (Reuse, 2012).

CECs can undergo biogeochemical transformations during in-stream natural attenuation processes (e.g., hydrolysis, oxidation, hydroxylation, conjugation, cleavage, de-alkylation, methylation, and demethylation) (B. Chen et al., 2009; Radke, Ulrich, Wurm, & Kunkel, 2010; Reuse, 2012). Attenuation of contaminants after wastewater discharge can vary widely as a function of the distance between discharge point and raw drinking water withdrawal (i.e., retention time), streamflow geometry (i.e., depth, mixing), and environmental conditions such as temperature, ultraviolet penetration, particulate matter, biological activity (Reuse, 2012). Survival of disease-causing microorganisms in the environment is of great concern and typical pathogen survival time (in days) at 20-30^{o}C in freshwater and wastewater are < 60 days (but usually < 30 days for Salmonella spp.); and < 30 days (but usually < 15 days for protozoa) (Metcalf et al., 2007).

Health-based violations. Under the enforcement and compliance of SDWA standards, health-related violations at public water systems are classified into Maximum Contaminant Level (MCL), Maximum Residual Disinfectant Level (MRDL), and Treatment Technique Requirement (TTR) (USEPA, 1996b). A treatment violation is a failure to properly treat a drinking water source to reduce the level of a specified contaminant. The studies on statistics of SDWA violations indicated that smaller CWSs tend to have more health-related violations than larger CWSs (Rubin, 2013; Wallsten & Kosec, 2008). A national assessment on all sized PWSs by Wallsten and Kosec (2008) indicated that smaller water systems were likely than larger water systems to have more MCL violations, and large PWSs may have a greater capacity to meet drinking water regulatory compliance. Rubin (2013) summarized statistics of violations for all the U.S community water systems (CWSs) which are year-round population serving PWSs and under all SDWA regulations. The findings reported that Disinfectants/Disinfection Byproducts Rule (D/DBPR) violations were likely to occur at smaller CWSs (serving between 501 and 10,000 people) than at larger CWSs. Figure 2.8 shows the information on health-related violations and treatment processes at DWTPs was obtained for the first quarter of the year 2020 (USEPA, 2019c). The smaller DWTPs had more health-based violations than the larger DWTPs and 32.5% of all the DWTPs in the U.S currently commit his violation.

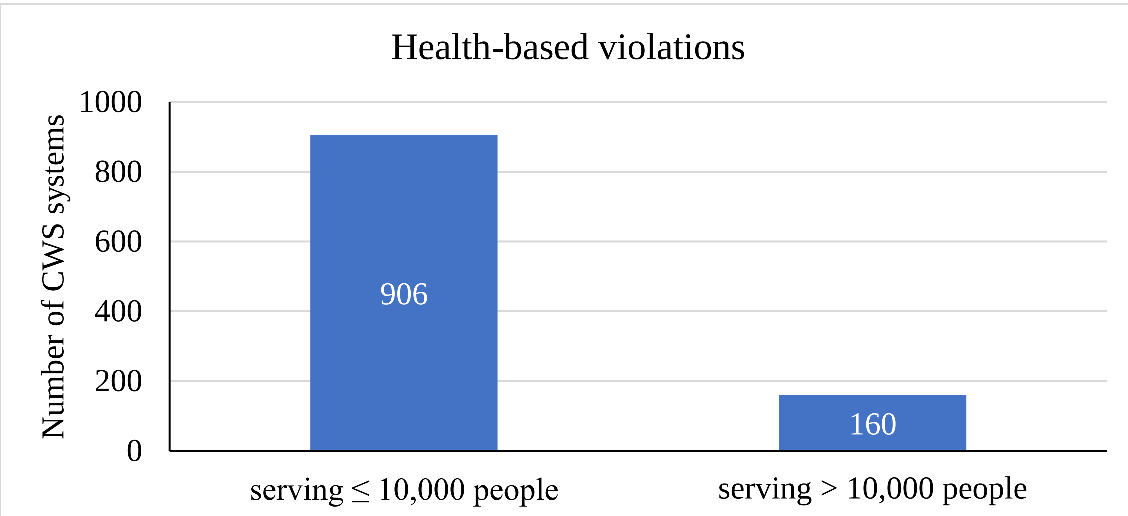

Figure 2.8. The number of Community Water Systems using Surface Water Violated Health-based Violations in the First Quarter of the Year 2020

The important factors of QMRA, level of DFR, and proximity analysis of contaminants (retention time, residence time) in surface water supplies, can be determined from the modernized DRINCS model. Thus, the results of this research can be used to fill the knowledge gap in epidemiological and risk assessment studies. Also, the DRINCS results can support the implementation of direct potable reuse schemes (blending advanced treated wastewater without surface water sources) that may still offer significant reductions in public health risks.

Relationship Between Water Quality and Human Health Risks

The Chemical Abstracts Services reported that nearly 90 million organic and inorganic chemicals have been registered in the USA, of which two-thirds are in commercial and approximately the addition of 15,000 new chemicals per day (Snyder, 2014).

The Safe Drinking Water Act (SDWA) was originally passed by Congress in 1974 to protect public health by regulating the nation's public drinking water supply. Treated wastewater discharges are a source of chemical and microbial pollution into surface water (USEPA, 1996b). Detection of CECs in waters has been well-documented such as their presence in effluent-impacted streams, finished water, and tap water (Benotti, Stanford, & Snyder, 2010; Benotti et al., 2009; Kolpin et al., 2002; T. Nguyen et al., 2018). Some constituents, such as microbial pathogens and contaminants of emerging concerns have the potential to affect human health, depending on their concentration, the routes/pathways, and the duration of exposure (Reuse, 2012). Pathogenic microorganisms are a particular focus because of their acute human health effects, and viruses necessitate special attention based on their low infectious dose, small size, and resistance to disinfection.

Impact on human health. CECs such as pharmaceutical active compounds (PhACs) and endocrine-disrupting compounds (EDCs) in waters are likely to have adverse biological effects on health at part per trillion concentrations (Weber, Khan, & Hollender, 2006). Long term exposure is associated with cancer, kidney damage, or nervous system problems (Peto, Gray, Brantom, & Grasso, 1991). *Cryptosporidium*

(Crypto) and human noroviruses (NoV) are two important waterborne pathogens commonly found in wastewater which are among the top 15 pathogens for the highest level of acute gastrointestinal illness (AGI) in the U.S. (Trussell et al., 2013). *Norovirus* (NoV virus), *Salmonella* (bacteria), *Cryptosporidium* (protozoan parasite) have caused a significant portion of waterborne illnesses in the U.S. Among the enteric viruses, NoV is the number one cause of AGI in the U.S that attributed to more than 20 million episodes and nearly 600 deaths in 2006 (Scallan et al., 2011). Past studies have assessed human health risks when exposing to waterborne pathogens such as Crypto, NoV, and Salmonella from direct ingestion of drinking water associated with de facto reuse (Amoueyan, Ahmad, Eisenberg, Pecson, & Gerrity, 2017; Chaudhry, Hamilton, Haas, & Nelson, 2017; Lim, Wu, & Jiang, 2017; Mac Kenzie et al., 1994).

A study by Mac Kenzie et al. (1994) documented the survival of pathogen *Cryptosporidium* oocysts through the filtration system in a water treatment plant in southern Milwaukee has caused the outbreak of acute watery diarrhea among more than 400,000 customers. The Milwaukee water treatment plant used surface water from Lake Michigan which received treated wastewater from upstream WWTPs. *Crypto, NoV, and Salmonella* were chosen as representative reference pathogens in a study by (Chaudhry et al., 2017). Knowledge of DFR at all DWTPS across the U.S will improve our understanding of DWTP treatment capability and the need to remove CECs in the water supply.

Qualitative microbial risk assessment (QMRA) in several studies has also evaluated the impact of retention time in the environmental buffers on pathogen die-off (Amoueyan et al., 2017; Lim et al., 2017). Pathogen decay is calculated based on pathogen-specific decay coefficient and wastewater effluent residence time which includes the in-stream traveling time; and residence time in a lake (if possible) from treated effluent outfall to surface water intake by a DWTP. The residence time in the lake can vary from 270 days to up to one year before withdrawal for drinking water treatment (Reuse, 2012). De facto reuse scenarios exhibited a higher annual risk of infection than planned potable reuse systems in varied traveling time of wastewater-derived pathogens. A case study of the Trinity River in Texas used a mean storage time of 270 days for Lake Livingston (Lim et al., 2017); a critical condition of 105 days at a temperature of 10 Celsius degree (Amoueyan et al., 2017). Also, the state of Massachusetts required the outfall of sewage (untreated wastewater) discharges to rivers was located more than 20 miles (32 km) upstream of drinking water intake (Reuse, 2012). Proximity Analysis is used to determine the relationship (measure the impact of multiple treated wastewater discharges) between treated wastewater outfalls (upstream WWTPs) which are point sources of CECs to drinking water supply source (DWTP) (Reuse, 2012). The DRINCS model is capable of estimating travel times which are important to understand the natural attenuation capacity of surface water systems for wastewater-derived CECs. While numerous case studies exist, there is a need to better quantity wastewater contribution of flow, CECs, and pathogens to downstream DWTPs of all sizes. Thus, the results of this research can be used to fill the knowledge gap in epidemiological and risk assessment studies.

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CHAPTER 3

MODELED DE FACTO REUSE AND CONTAMINANTS OF EMERGING CONCERN IN DRINKING WATER SOURCE WATERS

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Abstract

De facto reuse is the percentage of drinking water treatment plant (DWTP) intake potentially composed of effluent discharged from upstream wastewater treatment plants (WWTPs). Results from grab samples and a De Facto Reuse in our Nation's Consumable Supply (DRINCS) geospatial watershed model were used to quantify contaminants of emerging concern (CECs) concentrations at DWTP intakes to qualitatively compare exposure risks obtained by the two approaches. Between nine and 71 CECs were detected in grab samples. The number of upstream WWTP discharges ranged from 0 to >1,000; comparative de facto reuse results from DRINCS ranged from <0.1 to 13% during average flow and >80% during lower streamflows. Correlation between chemicals detected and DRINCS modeling results were observed, particularly DWTPs withdrawing from midsize water bodies. This comparison advances the utility of DRINCS to identify locations of DWTPs for future CEC sampling and treatment technology testing.

Keywords: *contaminants of emerging concern, de facto reuse, drinking water source water, transport, and fate*

Raw drinking water supplies are commonly under the influence of treated wastewater discharged upstream of drinking water treatment plant (DWTP) surface water intakes, a situation identified as unplanned or unintentional indirect potable reuse (i.e., de facto reuse [DFR]). For drinking water supplies serving more than 10,000 people from surface water sources, roughly half of these facilities in the United States are affected by at least one upstream treated wastewater discharge, based on previous geographical information systems (GIS)-based modeling efforts (Rice & Westerhoff, 2015, 2017; Rice, Wutich, & Westerhoff, 2013). Concurrently, studies of drinking water sources have detected pharmaceuticals and other contaminants of emerging concern (CECs) of wastewater origin when these sources are located downstream of treated wastewater discharge locations (Bradley et al., 2017; Glassmeyer et al., 2005). Although environmental sampling is the only sure means to identify and quantify contaminants present in a given waterbody, such monitoring campaigns can be costly. The interpretations of the results, which typically reflect specific conditions at a fixed point in time, are complicated by daily or seasonal differences in wastewater discharge flow or natural hydrologic streamflow variations in the rivers between the points of wastewater discharge and downstream drinking water intake. In theory, tens of thousands of CECs of wastewater origin and their corresponding transformation products could be monitored in water, and multiple preferred lists of approximately 1,000 target or surrogate compounds have been identified as indicators of wastewater in rivers (Bradley et al., 2017; Dickenson, Drewes, Sedlak, Wert, & Snyder, 2009; Dickenson, Snyder, Sedlak, & Drewes, 2011; Kolpin et al., 2002; Mawhinney, Young, Vanderford, Borch, & Snyder, 2011). Chemical mixture complexity is further amplified when multiple wastewater treatment plants (WWTPs) discharge into the same watershed that provides water to downstream drinking water intakes.

To better understand a drinking water utility's potential contribution to human or ecological exposure to organic CECs of wastewater origin under a range of streamflow conditions, a model (De Facto Reuse in our Nation's Consumable Supply DRINCS) has been developed to estimate the DFR across the United States (Rice & Westerhoff, 2015, 2017; Rice et al., 2013). Herein we compare results from a specific sampling effort analyzing surface water intakes from 22 surface water treatment plants for 192 organic

CECs, with pre-dictions of DFR from DRINCS. The relative location and distance of WWTP discharge points upstream of DWTP intakes are presented, along with the design capacity of the WWTPs, to aid in the comparison and interpretation of the model and chemical results. The objective is to increase the understanding of how the proximity of upstream WWTP discharges increases the vulnerability of downstream surface water DWTPs to contaminants of wastewater origin across the United States.

Methods

Water treatment plant selection and CEC monitoring information. CEC occurrence information for source water and treated waters at DWTPs were previously reported by Glassmeyer et al. (2017). This study uses the data from Phase II of Glassmeyer et al. (2017) for the 192 organic CECs from the 22 surfaces water DWTPs.

A detailed description of the criteria used to select sampling sites, sample collection procedures, analysis methods, and quality assurance and control protocols have been previously published Glassmeyer et al. (2017). In summary, intake grab samples from DWTPs were collected by personnel at participating DWTPs. Samples were packed on ice and shipped overnight to their destination laboratories at the US Environmental Protection Agency (USEPA) or US Geological Survey (USGS) for analysis. All methods have been described previously (Batt, Kostich, & Lazorchak, 2008; Boone et al., 2014; Cahill, Furlong, Burkhardt, Kolpin, & Anderson, 2004; Conley et al., 2017; Furlong et al., 2014; Furlong, Werner, Anderson, & Cahill, 2008; Schultz & Furlong, 2008; Ternes et al., 2005; USEPA, 1994, 2001, 2005; Zaugg, Smith, Schroeder, Barber, & Burkhardt, 2006).

Qualitative versus quantitative detections. Quantitative CEC concentrations and qualitative detection frequencies were previously reported (Glassmeyer et al., 2017). Samples that did not exceed their associated minimum reporting concentration—whether the lowest concentration minimum reporting level or reporting limit—but were above the instrument detection limit were considered a CEC qualitative detection (Glassmeyer et al., 2017). Additionally, samples with associated laboratory fortified matrix samples with >150% recovery were also considered qualitative detections. For both sub detection limit and matrix enhancement scenarios, we were sufficiently uncertain of the actual concentration that we did not report quantitative concentrations. "Qualitative detection frequency" used in the tables and figures in this study includes the detection limit and matrix enhancement censored analytes as well as the quantitatively reported analytes (i.e., every analyte that can be considered a positive detection). Quantitative detection frequency includes only those analyte detections with a concentration that can be reported with analytical certainty.

DRINCS model. The DRINCS model, previously developed and validated by Rice et al. (2013) Rice and Westerhoff (2015) and Rice, Via, and Westerhoff (2015), is a GIS model that incorporates spatially resolved data layers on the national hydrologic network. The outputs of DRINCS are calculated values of DFR linked with geospatial locations that can be mapped. Discharge locations of treated wastewater from WWTPs and sampling sites of the surface water source at DWTPs were used to estimate DFR at drinking water intake under average streamflow conditions. Spatial hydrography data in the United States were obtained from the National Hydrography Dataset Plus (NHDPlus, 2012; Viger, Rea, Simley, & Hanson, 2016), which represents the nation's drainage networks and related features, including rivers, streams, canals, lakes, ponds, glaciers, coastlines, dams, and USGS stream gauge data. USGS stream gauge attribute data include average, minimum, maximum, and percentile streamflows. The statistical values were calculated based on the entire record period ending Apr. 20, 2004, which is the end date for the NHD database employed (USEPA, 2007).

The WWTP locations and attribute data were obtained from the Clean Watershed Needs Survey 2008 (USEPA, 2008), which included 15,837 municipal WWTPs in the United States; we included the facilities (n= 14,651) that currently discharge to surface waters. Supporting attribute data for WWTPs included facility name, National Pollutant Discharge Elimination System (NPDES) permit number, level of treatment (primary, secondary, and tertiary), and present design flow. The level of treatment of effluent at WWTPs is a driver determining the potential occurrence of CECs, as concentrations substantially decrease with higher levels of treatment. Our analysis of treated municipal wastewater discharges from WWTPs included combined sewer systems but did not take into consideration combined sewer overflows or wet-weather bypasses (both of which can yield significant CEC loads; Phillips et al. (2012), or non-WWTP entities with NPDES permits to discharge. Conservative, and possibly worst-case, assumptions in calculating DFR made in the previous study (Rice et al., 2013) were used and include (1) WWTP discharge was equal to that of the present design capacity; (2) WWTP effluent had no in-stream loss; and (3) all water bodies were completely mixed. A Python program automated the process performed in the previous study (Rice et al., 2013). Levels of DFR were calculated from the cumulative upstream WWTP design discharges ($\Sigma Q_{ww,\,i}$) divided by the streamflow at the surface water DWTP intake (Q_{SW}):

$$\mathrm{DFR} = \frac{\sum Q_{ww,i}}{Q_{sw}} * 100\% \qquad (1)$$

Network relationships in streams between upstream WWTPs and each receiving DWTP at a regional scale were derived using a computerized mapping and analytics platform. Flow direction was established using the digitized direction from an attribute table of the stream network (NHD). The tracked upstream streamflow obtained from the geometric network was used to build a network analysis of routes (waterways). Using network analyst tools, proximal river distances from WWTPs upstream to each DWTP were determined by using the closest facility function based on Dijkstra's algorithm (Allen & Coffey, 2009; Dijkstra, 1959). Before use at all of the DWTP intakes, the Dijkstra algorithm approach was validated against the Ruler tool in Google Earth.

Results

Detection of CECs at DWTP intakes. As previously reported (Glassmeyer et al., 2017), the number of qualitatively detected analytes in the source water ranged from 30 in DWTP 29 to a maximum of 104 in DWTP4. Excluding detection frequency for microbial and inorganic chemicals we are not considering in this analysis, the number of the 192 organics CECs qualitatively detected ranged from nine in DWTP 5 to 71 in DWTP 4.

Magnitude and factors influencing DFR levels at DWTPs. ***DFR under mean streamflow.*** The DRINCS predicted levels of DFR ranged from 0 to 12.8% under mean streamflow (Appendix B). Three DWTPs (13, 23, and 29) on surface waters had no predicted wastewater impacts, as there were no upstream WWTP discharges identified within the watershed. Three DWTPs (5, 12, and 24) were groundwater systems, and DFR values were not predicted for these. Nineteen of the 22 DWTPs in Phase II with surface water sources had at least one upstream WWTP discharge, and DFR levels could be predicted by DRINCS. These DWTPs will be the focus in the remaining part of this section.

Streamflow and Strahler stream order. Impacts of varying streamflow (daily, seasonal, and annual) were considered two ways in the DRINCS model. First, historical streamflow data were used to obtain fifth and 90th percentile streamflows because these influences the potential range of higher to lower DFR values, respectively, that could be expected to occur at a DWTP intake. Second, source waters were classified based on the Strahler stream order (Strahler, 1957). For this classification, each segment of a river within a watershed can be considered as a node in a tree. When two first-order streams come together, they form a second-order stream, two second-order streams must flow together to form a third-order stream, and soon. Streams of lower order joining a higher order stream do not change the order of the higher stream. With two exceptions, the surface water DWTPs in this study are located on fifth- to ninth-order streams (Table 3.1).

Figure 3.1 summarizes these ranges in streamflow (part A) and associated DFR for DWTPs (part B), further classified based upon Strahler stream order. Generally, higher stream orders have higher mean flow rates (Figure 3.1, part A). The ratio of 90th:5th percentile streamflows (Q90/Q5) provides a relative indicator for the potential variation in streamflow. Table 3.1 summarizes Q90/Q5 ratio values, which range from 4 to 36. Higher streamflow dilutes wastewater discharged upstream of DWTP intakes. Variations in streamflow are generally seasonal and larger than variations in discharge flow rate from WWTPs, which fluctuate diurnally and are designed with peak hourly discharge to average daily discharge ratios of between 2 and 4. Whereas wet-weather wastewater discharges can be quite variable, the difference between winter and summer wastewater discharge flow rates is generally less than a factor of 2. Thus, variations in streamflow would be expected to alter DFR (Eq 1) to a larger extent than variations in wastewater discharge flow rates.

DFR values are shown in Figure 3.1, part B, on a log scale, with the open circle representing DFR at median streamflow; DFR at mean streamflow is listed in Table 3.1. Higher DFR values occur at lower streamflows (Eq 1). Although exceptions occur, higher Strahler stream orders have lower DFR values (Figure 3.1, part B). The highest predicted DFR under low flow conditions is 84% at DWTP 4 (Figure 3.1, part B); the mean DFR is listed in Table 3.1. The highest maximum DFR values are for DWTPs located on fifth- and sixth-order streams (Figure 3.1, part B).

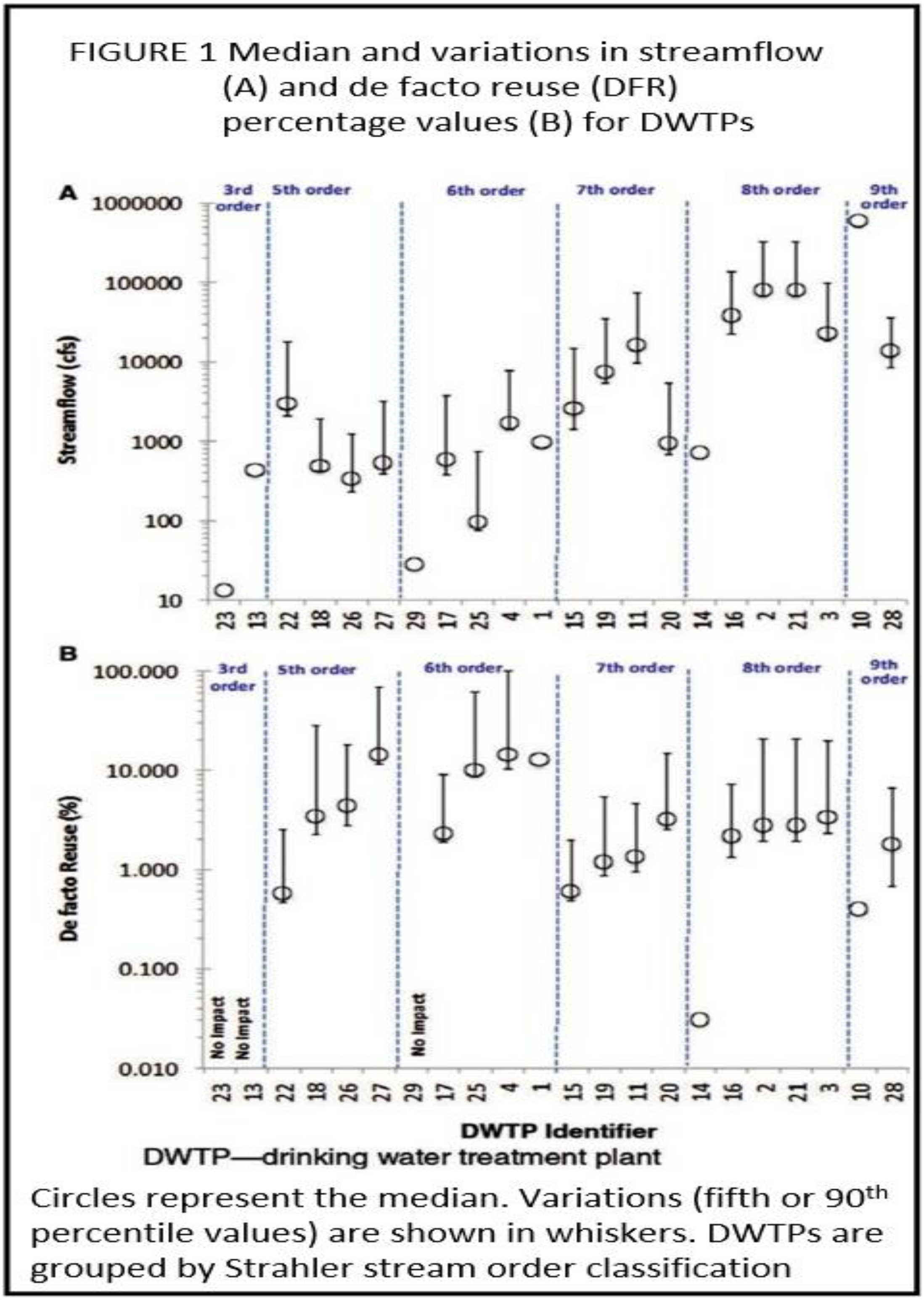


Figure 3.1 Median and Variations in Streamflow (A) and De Facto Reuse (DFR) Percentage Values (B) for DWTPs

Table 3.1. Geographical and Hydrological Information on Sampled Locations

DWTP #	Region of the United States	Water Source	Stream Order	Q90: Q5 Ratio	Reported Relative Water Level at Sampling	Annual Mean Streamflow at DWTP Intake cfs	De Facto Reuse at Mean Streamflow %
DWTP 01	South Central	River	6	NA	Average	966	12.8
DWTP 02	North Central	River	8	21	Slightly low	95,353	2.2
DWTP 03	North East	River	8	16	Average	32,861	2.3
DWTP 04	North East	River	6	21	Average	3,026	7.8
DWTP 05	South West	Groundwater	—	—	—	—	—
DWTP 10	South Central	River	9	NA	Low	587,823	0.4
DWTP 11	North West	River	7	9	Above average	27,399	0.8
DWTP 12	North Central	Groundwater	—	—	—	—	—
DWTP 13	North East	Reservoir	3	12.3	Average	419	0
DWTP 14	South East	Lake	8	NA	Average	718	0.03
DWTP 15	Plains	River	7	11	Average	4,498	0.3
DWTP 16	Plains	River	8	6	High	30,980	2.7
DWTP 17	South East	River	6	15	Above average	1,056	1.3
DWTP 18	North East	River	5	20	Average	644	2.6
DWTP 19	Plains	River	7	14	Average	8,581	1
DWTP 20	South West	River	7	16	Average	2,006	1.5
DWTP 21	North Central	River	8	21	Average	95,353	2.2
DWTP 22	South East	River	5	16	Average	1,817	0.9
DWTP 23	South East	Reservoir	3	13	Low	16	0
DWTP 24	Plains	Groundwater	—	—	—	—	—
DWTP 25	Plains	Reservoir	6	36	Very low	213	4.3
DWTP 26	North East	River	5	8	Average	527	2.8
DWTP 27	North Central	River	5	19	Low	1,177	6.4
DWTP 28	South West	Reservoir	9	4	Low	16,635	1.4
DWTP 29	South Central	Reservoir	6	NA	Average	28	0

DWTP—drinking water treatment plant, NA–not applicable, Q90:Q5–the ratio of 90th:5th percentile streamflows
Dashed lines indicate groundwater locations therefore excluded from calculation.

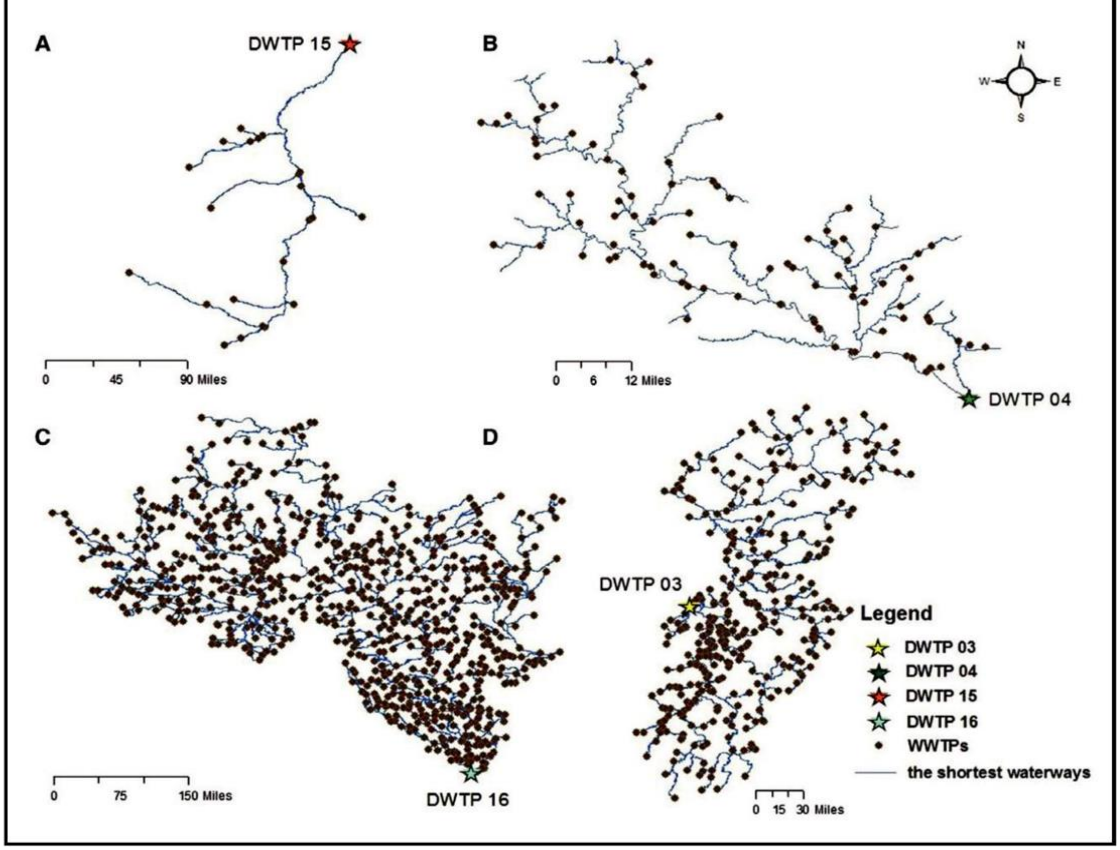


FIGURE 2 Illustration of watersheds for four drinking water treatment plants (DWTPs) showing rivers (blue lines), location of multiple wastewater treatment plant (WWTP) discharge locations (solid circles), and locations of DWTPs (star symbols) that represent the downstream end (lower elevation) of the watersheds

Figure 3.2 Illustration of the Watershed for Four Drinking Water Treatment Plants (DWTPs) showing Rivers (Blue Lines), Location of Multiple Wastewater Treatment Plant (WWTP) Discharge Locations (Solid Circles), and Locations of DWTPs (Star Symbols) that Represent the Downstream End (Lower Elevation) of the Watershed

Number and size of upstream WWTP discharges. DWTPs experience different amounts of DFR based on the presence of upstream WWTPs. Table 3.2 provides information on the number and design treatment capacity of WWTPs located upstream of each DWTP intake. DWTP 2 and 21 (plants nearly co-located on the same river) have 1,200 upstream WWTPs that account for up to 1,372 mgd of treated wastewater, but 442 of these WWTPs are small and have design discharges below 0.1mgd. At an average daily sewage production of 75 gpd per person, a 0.1 mgd facility serves a population of roughly 1,300 people. In total, 3,615 (82%) out of 4,392 of the WWTPs upstream of the DWTPs in this study have design capacities below 1 mgd (Table 3.2). Only 59 out of nearly 4,392 WWTPs considered in this study have design capacities >10 mgd (Table 3.2). Thus, there are large numbers of small WWTPs in the studied watersheds.

Figure 3.3 Normalized Cumulative Distributions (F, Dimensionless) of WWTP Wastewater Flows, Relative to the Total Upstream WWTP Flow

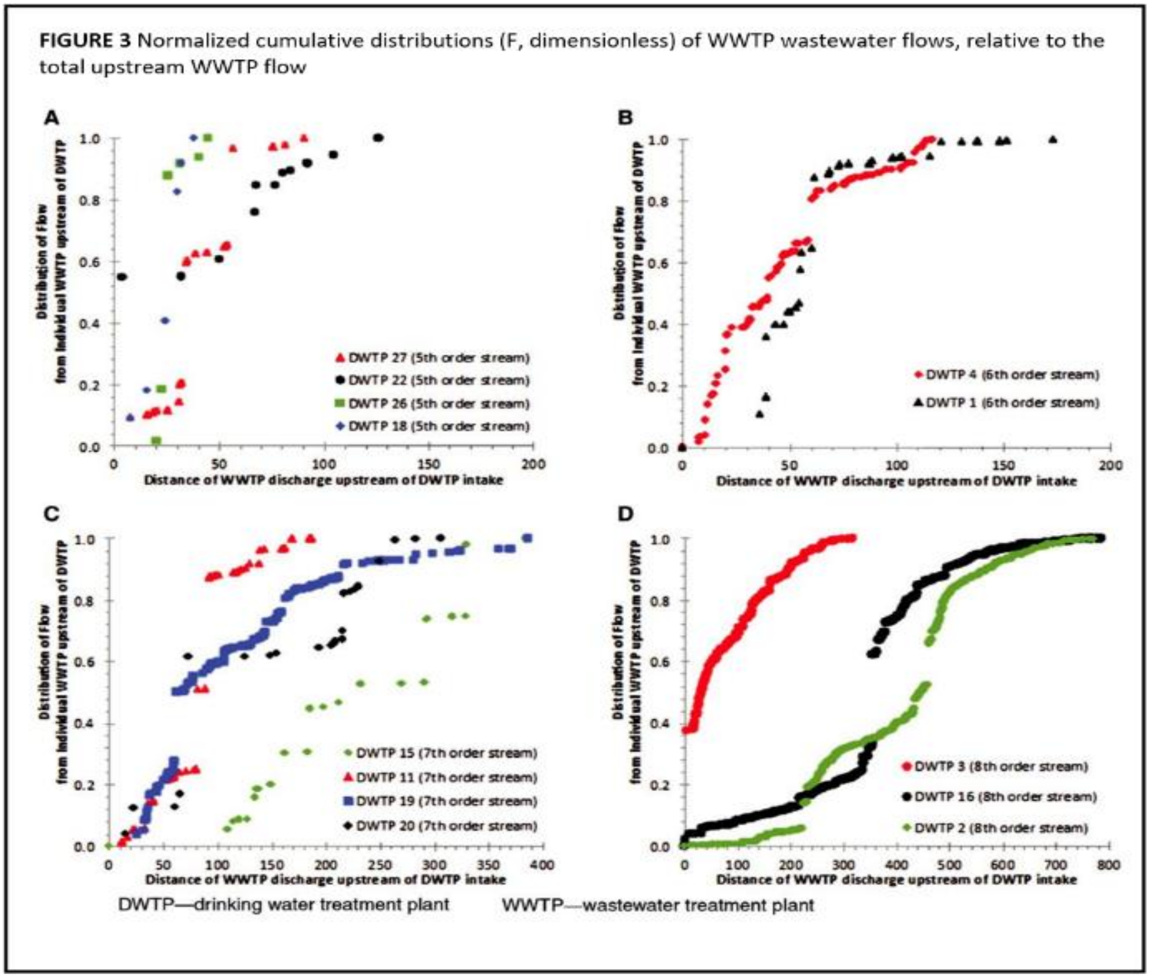


Table 3.2 Location and Sizes of Wastewater Treatment Plants Located Upstream of Drinking Water Treatment Plants Investigated in this Study

DWTP	Location of Upstream WWTPS		Size of Upstream WWTPs				Total Impact of Upstream WWTPs	
	Less than 10 mi	Maximum distance	Less than 0.1 mgd	0.1–1 mgd	1–10 mgd	Greater than 10 mgd	Total Upstream WWTPs	Accumulated Upstream WW Discharges $Q_{ww,T}$ mgd
#	n	mi	n	n	n	n	n	
DWTP 01	0	115	4	14	11	2	31	80
DWTP 02	1	771	442	529	215	14	1,200	1,372
DWTP 03	1	315	103	171	73	3	350	491
DWTP 04	3	116	16	42	34	1	93	153
DWTP 05	—	—	—	—	—	—	—	—
DWTP 10	6	905	178	212	67	8	465	1,459
DWTP 11	0	187	4	27	8	3	42	135
DWTP 12	—	—	—	—	—	—	—	—
DWTP 13	0	0	0	0	0	0	0	0
DWTP 14	0	54	0	1	0	0	1	0.15
DWTP 15	0	384	10	9	3	0	22	10
DWTP 16	3	785	388	326	60	9	783	534
DWTP 17	1	63	0	2	2	0	4	9
DWTP 18	1	38	0	4	2	0	6	11
DWTP 19	0	386	51	62	10	1	124	56
DWTP 20	0	306	7	11	5	0	23	19
DWTP 21	1	771	442	529	215	14	1,200	1,372
DWTP 22	1	126	3	6	2	0	11	11
DWTP 23	0	0	0	0	0	0	0	0
DWTP 24	—	—	—	—	—	—	—	—
DWTP 25	1	438	0	4	2	0	6	6
DWTP 26	0	45	0	4	2	0	6	9
DWTP 27	1	90	1	10	5	2	18	49
DWTP 28	0	660	1	2	2	2	7	155
DWTP 29	0	0	0	0	0	0	0	0

DWTP—drinking water treatment plant, WW—wastewater, WWTP—wastewater treatment plant
Dashes indicate groundwater locations therefore excluded from calculation.

The proximity of upstream WWTP discharges. DFR values were calculated from the cumulative upstream wastewater discharges (Eq 1), and do not directly account for the proximity between WWTP discharges and the DWTP intake. A single value to represent proximity was difficult to derive because of the complexity of watersheds. Figure 3.2 illustrates the watersheds and location of WWTP discharges upstream of four DWTP intakes (DWTPs 3, 4, 15, and 16). There are complex networks of tributaries in these watersheds, many with several WWTP discharges. Figure 3.3 documents the proximal distance between upstream WWTP discharge and the DWTP intake for several watersheds; each symbol represents a WWTP discharge.

The y-axis in Figure 3.3 represents the distribution of treated wastewater flows from upstream WWTPs (Qww, i), normalized to cumulative wastewater flow from all upstream WWTPs ($Q_{ww,\,T}$), separated by stream order. The value of $Q_{ww,\,T}$ varies by site and is summarized in the last column of Table 3.2. Eleven of the DWTPs (e.g., DWTPs 18 and 26 in Figure 3.3, part A) have WWTP discharges located within 10 mi upstream of a DWTP intake. In others, few individual WWTPs contribute substantially to the overall wastewater flows into the surface water source serving the DWTP, as indicated by breaks or jumps in the plot (e.g., DWTP 19 in Figure 3.3, part C, and DWTP 3 in Figure 3.3, part D). In other systems (e.g., DWTPs 2 and 16 in Figure 3.3, part D), most of the wastewater flow originates hundreds of miles upstream.

Several indicators were used to quantify the complex relationship between multiple WWTP discharge points located at multiple upstream locations. As illustrated in Figure 3.3, any DWTP location can be modeled as having a distribution of upstream sources. Several statistical functions can be used to fit such distributions and serve as indicators, but they typically require multiple fitted parameters. As discussed subsequently, the purpose of such indicators is to provide a secondary index of the relative risk of having CECs of wastewater origin in DWTP source supplies. Consequently, correlating multiple fitted parameters were not deemed appropriate. Two alternative indexes were developed to further examine the relation between WWTP locations and CEC detections. To calculate the first index, the magnitude ($Q_{ww,\, i}$ in mgd) and distance (M_i in mi) of each (i) WWTP located upstream were considered to be inversely proportional (Eq 2) to the relative risk of CEC occurrence in DWTP intakes; $Q_{WW,\, T}$ is the cumulative discharge of all upstream WWTPs ($Q_{WW,\, T} = \Sigma Q_{WW,\, i}$). A singular proximity index (PI) from all upstream WWTPs was then calculated by summing these values (Eq 3). Larger PI values suggest larger WWTP discharges located closer to DWTP intakes and could indicate a larger potential wastewater impact. For the second index, the relative skewness (SK) of the distribution functions illustrated in Figure 3.3 (F being the y-axis distribution from 0 to 1) were considered as potentially being useful indicators for differences among watersheds. A simple metric was used to quantify this SK. As shown in Eq 4, SK was related to the distance ($M_{0.1}$) associated with F = 0.1 divided by the distance ($M_{0.5}$) associated with F = 0.5 in Figure 3.3; F is the normalized cumulative distributions of WWTP wastewater flows (i.e., y-axis value in Figure 3.3). SK would range from 0 to 1. Higher SK values suggest larger WWTPs located closer to the DWTP. There is no direct relation between PI and SK, but each index can be used separately to compare proximity patterns among different DWTPs. PI and SK values are summarized in Table 3.3.

$$\mathrm{PI_i} = \frac{\mathrm{Q_{ww,i}}}{\mathrm{M_i}} \qquad (2)$$

$$PI = \frac{Q_{ww,i}/M_i}{Q_{ww,T}} * 1000 \quad (3)$$

$$SK = \frac{M_{0.1}}{M_{0.5}} \quad (4)$$

Qualitative comparison of CEC detection and DRINCS model DFR predictions. The over 4,000 WWTPs present in the watersheds of the DWTPs studied herein include a wide range of treatment processes from aerated lagoons to advanced nutrient control. Biodegradation, biosorption, volatilization, hydrolysis, oxidation, and other biochemical or physical processes within different types of WWTPs can potentially influence the extent of CEC removal. DRINCS does not directly account for these differences in the treatment process, and DFR simply represents a conservative estimate for the potential risk of having surface DWTP supplies containing CECs of wastewater origin. However, the variability of the WWTP unit process upon CEC removal is expected to affect absolute CEC concentrations present at downstream DWTP intakes. The flow of the water body affects the transport time for WWTP effluents to reach the down-stream DWTPs. At a velocity of 1 ft/s (0.3 m/s), the travel time is approximately one week for 100 mi; above 10 ft/s (3 m/s), the travel time is less than a day-one day for 100 mi.

CECs can continue to biodegrade in water after wastewater is discharged to rivers and before entering DWTPs. Additional biogeochemical processes (adsorption to sediment, volatilization, photolysis, and so on) can also occur in rivers (Chen, Lee, Westerhoff, Krasner, & Herckes, 2010; Chen, Nam, Westerhoff, Krasner, & Amy, 2009; Chen, Westerhoff, & Krasner, 2008; Karanfil, Krasner, Westerhoff, & Xie, 2008). These time-dependent processes depend on other water quality factors (temperature, pH, turbidity, and so on), location, river depth, and others. The CECs within this data set will have a range of persistence both during wastewater treatment and environmental transport (Glassmeyer et al., 2005). In general, the per- and poly-fluoroalkyl substances (PFASs) are more resistant to treatment (Rahman, Peldszus, & Anderson, 2014; Zhang, Yan, Li, & Zhou, 2015) and more stable in the environment (Happonen et al., 2016; Nguyen et al., 2017; Wang, Wang, Zhao, Cao, & Wan, 2015) than most CECs.

Table 3.3 Results of Modeling and Summary of Chemical Analyses of Drinking Water Source Water Samples

DWTP #	De Facto Reuse at Mean Streamflow %	Proximity Index (PI)	Skewness Index (SK)	Number of Organic Chemicals Qualitatively Detected (n = 192)	Number of Organic Chemicals Quantitatively Detected (n = 192)	Sum Quantitative Detections ng/L
DWTP 01	12.8	20	0.67	31	16	135.3
DWTP 02	2.2	3.5	0.57	32	18	1,075.20
DWTP 03	2.3	169	1	55	31	820.2
DWTP 04	7.8	40	0.26	71	35	1,425.50
DWTP 05	—	—	—	9	4	27.5
DWTP 06	0.4	43	0.02	28	18	460.9
DWTP 07	0.8	16	0.41	15	6	1.5
DWTP 08	—	—	—	18	11	69.9
DWTP 09	0	0	0	12	9	3.3
DWTP 10	0.03	19	1	13	8	2.7
DWTP 11	0.3	4.9	0.69	14	6	1.5
DWTP 12	2.7	30	0.42	15	13	350.1
DWTP 13	1.3	39	1	26	13	293.4
DWTP 14	2.6	46	0.24	26	22	265.6
DWTP 15	1	15	0.5	24	10	27.6
DWTP 16	1.5	15	0.31	22	8	20
DWTP 17	2.2	3.5	0.86	36	20	877.6
DWTP 18	0.9	156	1	37	26	1,762.20
DWTP 19	0	0	0	18	11	138.3
DWTP 20	—	—	—	13	11	541.7
DWTP 21	4.3	64	1	19	11	17.7
DWTP 22	2.8	39	0.84	44	20	451.9
DWTP 23	6.4	35	0.44	41	24	1,699.70
DWTP 24	1.4	39	1	24	15	232.4
DWTP 25	0	0	0	15	8	31.1

Concentration data source: Glassmeyer et al. 2017.
DWTP—drinking water treatment plant
Dashes indicate groundwater locations therefore excluded from the calculation.

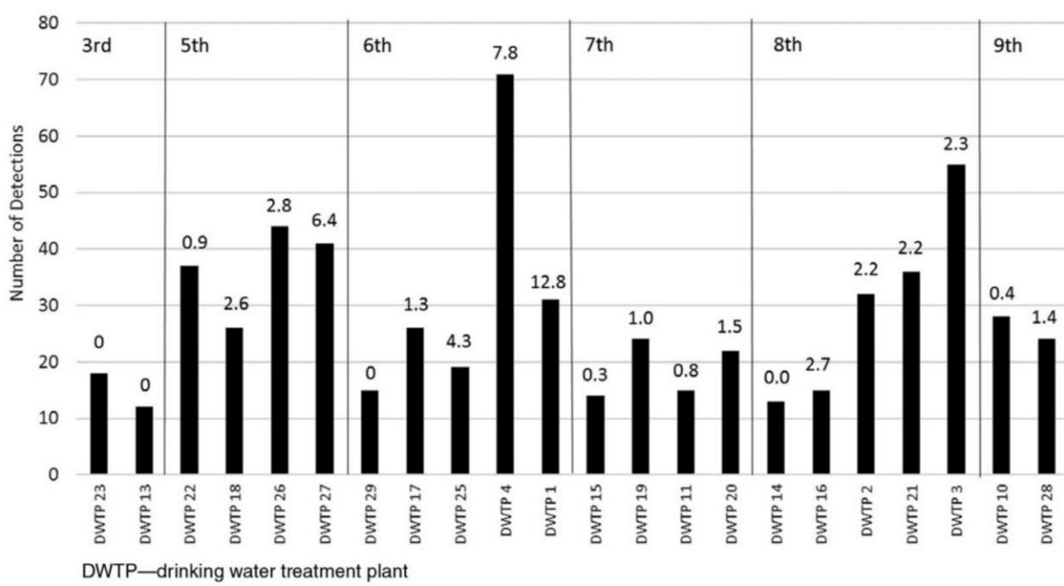
The CEC source water data set is comprehensive both in terms of the number of chemicals analyzed and the number of DWTPs sampled (n= 25). However, grab samples are only representative of a single point in time, many of the CECs were below reporting or detection limits, and quantitative concentrations were not reported. Therefore, the researchers made a qualitative comparison of CEC occurrence and DRINCS model outputs rather than using formal statistical analysis. Table 3.4 provides a cursory comparison of the field and modeling potential for CECs to occur at DWTP intakes. The general trend is that more CECs are detected at higher DFR values.

Figure 3.4 displays the number of qualitative detections for all of the CECs at each location. In general, within each stream order, the number of detected analytes increases as DFR increases. One notable exception to this trend is DWTP 1. The field blanks associated with this location had measurable concentrations of many commonly detected analytes, such as atrazine, caffeine, cotinine, meprobamate, coprostanol, galaxolide, N,N-diethyl-meta-toluamide, tri(2-butoxyethyl) phosphate, and tri (2-chloroethyl) phosphate. Per our quality assurance/quality control protocol (Glassmeyer et al., 2017) (Glassmeyer et al. 2017), the detections of these analytes in the DWTP 1 samples were censored, as the concentrations in the samples needed to exceed blank detections by a factor of three or more to be retained. Without the removal of the field blank censored detections, DWTP 1 would have more measured detections, and a general trend of increasing detections with increasing DFR within a stream order would hold.

Table 3.4 Comparison Between De Facto Reuse (DFR) and Number of Contaminants (CECS) Qualitatively Detected at Drinking Water Treatment Plant (DWTP) Intake

No. of Qualitative Detections of CECs at DWTP Intakes	**Level of DFR Determined Under Mean Flow in DRINCS**				
	Not Impacted	**<0.1%**	**0.1 - 1%**	**1- 5%**	**> 5%**
<10	-	-	-	-	-
10 - 20	2	1	2	3	-
20 - 30	-	-	1	5	-
30 - 40	-	-	1	2	1
>40	-	-	-	2	2

FIGURE 4 Number of organic contaminant of emerging concern analytes detected at each sampled location, sorted by stream order



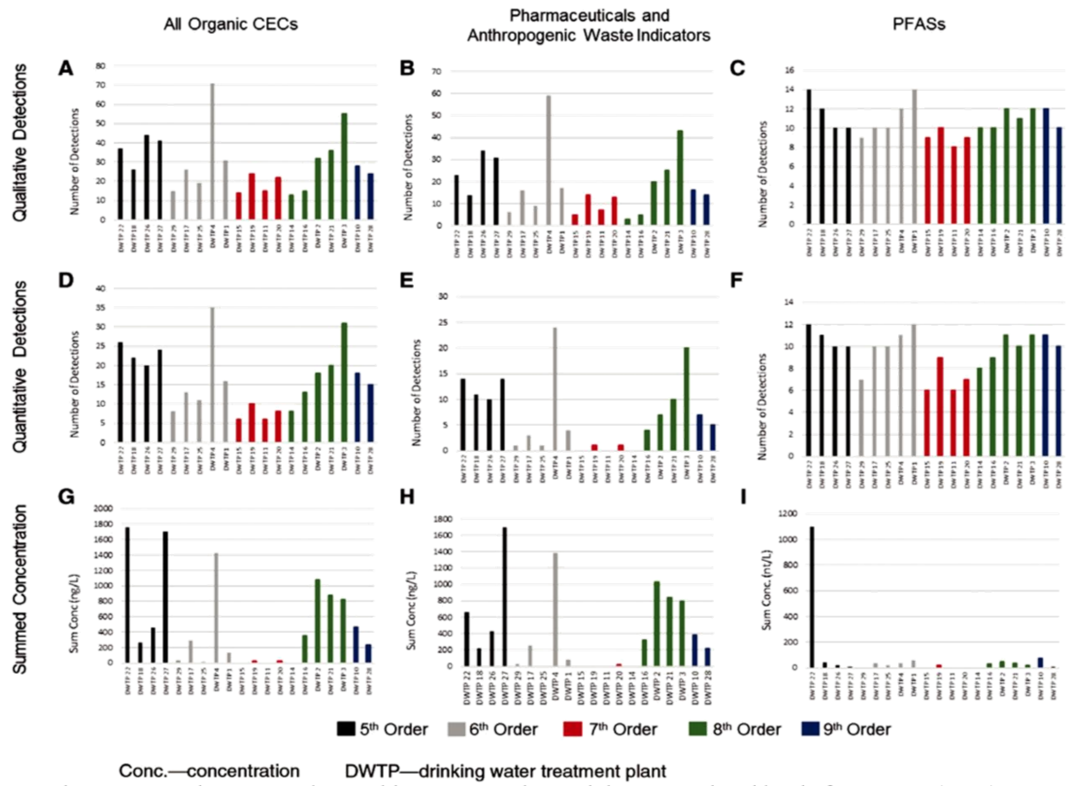
The order of the DWTPs reflects the stream order and de facto reuse (DFR) percentage at median streamflow from Figure 1. The vertical lines on the graph separate the stream orders. The number above each bar is the DFR at mean streamflow (Table 1).

Figure 3.4. Number of the Organic Contaminant of Emerging Concern Analytes Detected at Each Sampled Location, Sorted By Stream Order

Figure 3.5 explores the trends between measured detections and stream order, and Figure 3.6 examines the relationship between concentration and DFR (parts A–C), PI (parts D–F), and SK (parts G–I). This figure examines all of the organic CECs (for both Figures 3.5 and 3.6, parts A, D, and G) and separates the relatively ephemeral pharmaceuticals and anthropogenic waste indicators (AWIs) (parts B, E, and H) from the more persistent PFASs (parts C, F, and I). In terms of both the qualitative and quantitative detections (Figure 3.5, parts A to F), the PFASs (parts C and F) show substantially less variability between the locations than the combined pharmaceuticals and AWIs (parts B and E). It is interesting to note that once the PFASs are removed, the DWTPs with seventh-order stream sources tend to have lower numbers of detections than the sixth- or eighth-order sources (Figure 3.5, parts B and E). More research would be needed to determine if this is a nationwide trend or unique to this data set.

The cross-site similarity of the PFAS is diminished when concentration is considered (Figure 3.5, part I). DWTP had a greater total PFAS concentration compared with the other locations. This illustrates one of the weaknesses of the DRINCS model: although the wastewater composition of the source water is in general a good indicator for relative contamination, unique non-wastewater sources may be significant contributors of CECs upstream of source water intakes. Turning to the three indexes discussed earlier (DFR, PI, and SK; Figure 3.6), several relationships can be noted. Excluding the DWTP 1 outlier, DFR shows the strongest trend of the three indexes when plotted against the sum of concentrations for all organic CECs (Figure 3.6, part A) and pharmaceuticals and AWIs (part B); higher DFRs are generally correlated with greater concentrations of CECs in the source water.

FIGURE 5 Qualitative detections, quantitative detections, and summed concentration for all organic contaminants of emerging concern (CECs), separated by pharmaceuticals and anthropogenic waste indicators and per- and polyfluoroalkyl substances (PFASs)



Bar color separates the DWTPs by Strahler stream order, and they are ordered by de facto reuse (DFR) at median streamflow (Figure 1).

Figure 3.5 Qualitative Detections, Quantitative Detections, and Summed Concentration for all Organic Contaminants of Emerging Concern (CECs), Separated by Pharmaceuticals and Anthropogenic Waste Indicators and Per- and Poly-Fluoroalkyl Substances (PFASs)

When DWTP 1 is excluded from the regression, the R^2 increases from 0.0507 to 0.3004 for all organic CECs (Figure 3.6, part A), and from 0.1032 to 0.5159 for the pharmaceuticals and AWIs (part B). For the PFASs, the trend is minimized by the high concentrations at DWTP 22 ($R^2 = 0.0445$; Figure 3.6, part C), but even when that point is omitted, the trend is not as strong as it is for the other analytes ($R^2 = 0.1506$). Removing the DWTP 1 outlier similarly does not increase the relation ($R^2 = 0.017$). When PI is plotted against the sum of the three different concentration sets (Figure 3.6, parts D, E, and F), the trends (R^2 of 0.183, 0.0406, and 0.2417, respectively) are not as strong as DFR trends with the DWTP 1 outlier removed. DWTPs 3 and 22 are two points outside the rest of the field on all the organic CEC and pharmaceutical and AWI-only graphs (and are indicated in Figure 3.6, parts D and E). Both of these locations have a large percentage of their total wastewater load (~38 and 55%, respectively; Figure 3.3, parts D and A) from WWTPs within a 10 mi distance. The PI may have an application for determining DWTPs that have relatively stronger impacts from individual nearby WWTPs, whereas DFR is a better indicator of the general wastewater impact. The SK value shows no relationship under any of the organic CEC permutations (Figure 3.6, parts G, H, and I).

To further examine the effect DFR may have on analyte detections, Appendix B compiles the maximum concentration and DWTP with the maximum detection for all 62 organic CECs quantitatively detected at least once in the source water. Eleven of the 25 DWTPs had at least one maximum concentration for any analyte measured in its respective source water, but DWTP 4 is distinctive in this group. It has more than twice as many maximum concentration detections as any other DWTP (with the exception of DWTP 22 PFAS concentrations). Additionally, the detections at DWTP 4 are often the study maximum for organic CECs detected at many locations (as indicated by a >4% quantitative frequency); the other DWTPs often are the maximum detection because they are the only detection. DWTP 4 had the greatest DFR of the 11 DWTPs on this list at 7.8% (Table 3.3). To see how these maximum concentrations in source water compare with measured concentrations in wastewater-influenced locations, the maximum concentrations for 40 CECs also measured in Bradley et al. (2017) are listed in Appendix B, along with the calculated relationship between the source and wastewater-influenced concentrations. (An additional 80 analytes detected in Bradley et al. were also monitored in Glassmeyer et al. (2017), but they were not quantitatively detected in the source water samples and therefore excluded from the analysis.) The affected surface water sites sampled in Bradley et al. (2017) were chosen to reflect mixed-contaminant exposure profiles, including but not limited to wastewater effluent; they are not the same locations as Glassmeyer et al. (2017), but represent impacted locations.

FIGURE 6 De facto reuse (DFR) at mean streamflow, proximity index (PI), and skewness (SK) index in relation to the summed concentration for all organic contaminants of emerging concern (CECs), and separated by pharmaceuticals and anthropogenic waste indicators and per-and polylfluroalkyl substances (PFASs)

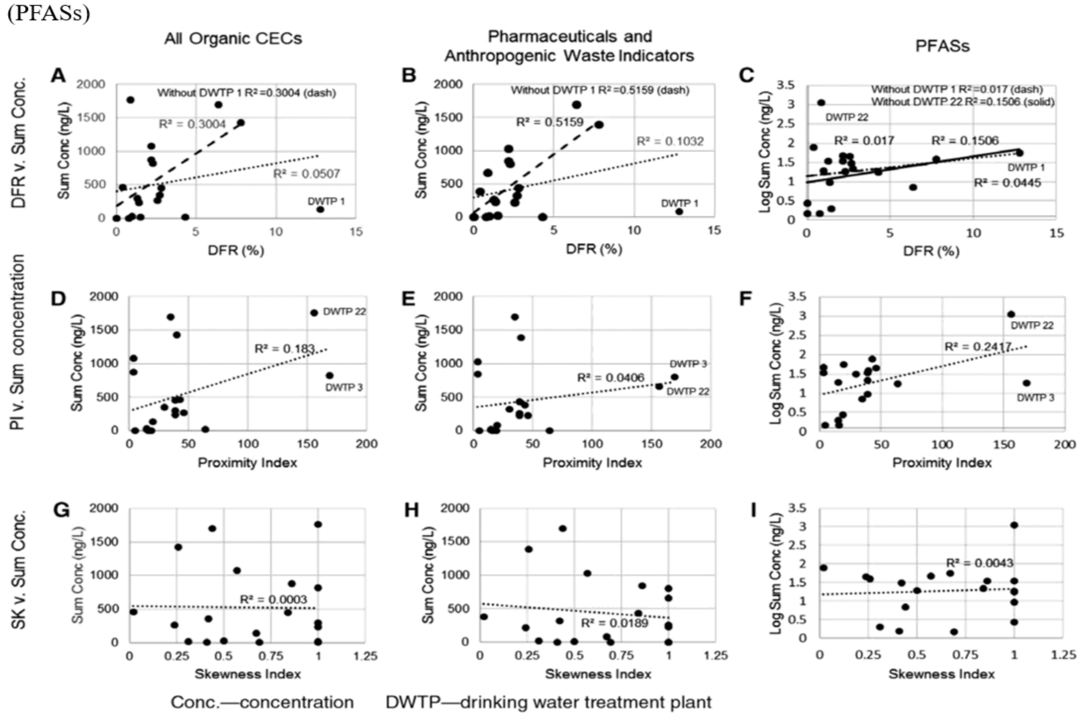


Figure 3.6 De Facto Reuse (DFR) A Mean Streamflow, Proximity Index (PI), And Skewness (SK) Index in Relation to the Summed Concentration For All Organic Contaminants of Emerging Concern (CECs), and Separated by Pharmaceuticals and Anthropogenic Waste Indicators and Per-And Polylfluroalkyl Substances (PFASs)

Of the 40 sources water maximum concentrations from Glassmeyer et al. (2017), 34 were <20% of the more wastewater-influenced sample maximum reported in Bradley et al. (2017) (Appendix B). The source water concentrations of cotinine, diltiazem, and desmethyl diltiazem were all between 25% and about 30% of the wastewater- influenced maximums, while verapamil, amitriptyline, and methyl-1H-benzotriazole were found in source waters at concentrations greater than the wastewater influenced locations (355, 118, and 130% relative concentration; Appendix B). The fact that 114 out of 120 contaminant pairs monitored in both studies were substantially lower in the drinking water source waters demonstrates the beneficial effect that dilution and other natural attenuation processes have on aquatic CEC concentrations. However, the six compounds with >25% relative concentration, and particularly the three components with higher concentrations (verapamil, amitriptyline, and methyl-1H-benzotriazole), illustrate the need for DWTPs to estimate and assess the potential for WWTP influence on the chemical contaminant composition of their source water.

DRINCS can identify situations in which potential exists for CECs in DWTP influents and assist with understanding the potential seasonal variability as a function of streamflow (Rice & Westerhoff, 2015, 2017). For DWTPs with higher levels of DFR under average flow (>1–5%), the frequency of CEC detection and CEC concentrations should be high enough to detect by modern analytical methods. Lower DFR values may have CECs that occur below current analytical detection limits (Rice et al., 2015). Thus, DRINCS emerges as a potentially useful tool to identify DWTPs at higher risk for CEC occurrence, where subsequent monitoring could be focused.

Conclusions

The variability in CEC detection at a particular DWTP intake depends on many factors including streamflow, type of treatment processes used at any upstream WWTP, WWTP discharge flow rates, travel distance, water quality within the receiving waters, and so on. As indicated in the prior study in which the CEC occurrence data were collected (Glassmeyer et al., 2017), the conclusion noted that samples collected at a single point in time make up a snapshot of occurrence, and future studies would benefit from more detailed and focused time series sample collection designs that better capture temporal variations. The general comparison of DRINCS and the “snapshot” of CEC occurrence data compared here advances the validity of using DRINCS as a tool to identify locations of DWTPs for future sampling and treatment technology testing. Before the development and simulation of the DRINCS model (Rice & Westerhoff, 2015), the only other available nationwide documentation linking drinking water sources to wastewater percentage was several decades old (Swayne, Boone, Bauer, & Lee, 1980).

Levels of DFR from DRINCS were previously compared with the potential occurrence of Unregulated Contaminant Monitoring Rule CECs (Rice & Westerhoff, 2015), which included only a few wastewater indicator compounds. However, this paper demonstrates, for the first time, the ability of DRINCS for a much broader range of CECs of wastewater origin and considers distances between WWTP discharges and DWTP intakes. Databases linked with DRINCS include populations served and type of unit processes at WWTPs and DWTPs; also, DRINCS can calculate the number, size, and proximity of WWTP discharges into surface waters upstream of DWTP intakes. Queries could be made that include some of the factors described herein that would affect CEC occurrence. Although the comparison of model and field results in this study indicates the general validity of the DRINCS model, the data also suggest that predictive capabilities could be enhanced by closer proximity of instream flow information, such as that provided by stream gage near DWTP intakes, to more accurately measure DFR. Ongoing improvements in chemical analytical capabilities and expansion of the range of CECs routinely determined will also serve to better anchor model predictions with observed ambient source water conditions.

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ENDNOTE

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CHAPTER 4

DEVELOPMENT OF GEOSPATIAL MODELING DRINCS 2.0

Introduction

A geospatial model, De facto Reuse Incidence in our Nation's Consumptive Supply (DRINCS), was previously developed and validated by Rice and Westerhoff (2015) to model the percentage of treated wastewater effluents present within downstream potable water supplies (i.e., de facto reuse). The DRINCS model is an ArcGIS-based model that is spatially explicit, with broad-scale capability, and an effective tool to support decision-making through the improved communication of GIS-based maps and visualization (Rice, Via, & Westerhoff, 2015; Rice, Wutich, & Westerhoff, 2013). This enables the model to couple with a Python script designed to perform network analysis on hydrologic regions across the U.S.

The DRINCS model is developed on a national scale and utilizes geospatial data and water utility information for public water systems using surface water (DWTPs) and wastewater treatment plants (WWTPs) (Rice et al., 2015). The primary large datasets include the Clean Watersheds Needs Survey, the National Hydrography Dataset Plus, and the EPA SDWIS federal database (Rice & Westerhoff, 2015). The EPA Safe Drinking Water Information (SDWIS) provides basic water system inventory such as population served, violations and enforcements, level of treatment, and information on the surface water source for public water systems (USEPA, 2019b). The National Hydrography Dataset Plus (NHDPlus), developed by the US EPA Office of Water and assisted by the US Geological Survey, is an integrated and application-ready geospatial database (NHDPlus, 2012). The NHD Plus based on the medium resolution NHD (1:100,000 scale) includes more than 3 million stream networks with value-added attributes. The EPA Clean Watersheds Needs Survey (CWNS) contains a basis WWTP facility information, design capacity, level of treatment, and wastewater outfalls to surface water (USEPA, 2012). When the model was created, the most current and available datasets in use were CWNS 2008 (released in 2010) for WWTP locations and flow data (N = 14,651), NHD PlusV1 (released in 2006), and NHD PlusV2 (released in 2012) for vector and attribute data, and the EPA SDWIS (in 2014) for DWTP locations and population served (N = 2,160). Since the conditions and the environment are changing, the accuracy of the model needs to be monitored. The periodical update of this primary database for the DRINCS model reflects the need for the model to be updated to keep up with all these changes.

Prior research indicated the limitations of the DRINCS model due to the location datasets (Rice et al., 2015). The geographical coordinates (latitude and longitude) of wastewater outfalls from WWTPs and surface water intakes by DWTPs, unfortunately, are often shown as a facility address rather than the exact locations along a river reach or water body. In these instances, much more effort was made by looking up coordinates, design capacity, and additional information on names of effluent receiving rivers, surface water sources (river, lake, canal, creek, reservoir, etc.) for each water system from other data sources. For example, the US EPA Enforcement and Compliance History Online (ECHO) is a good data source to fill in the data gaps (USEPA, 2020). Alternatively, the missing information was found through individual reports, for example, the National Pollutant Discharge Effluents (NPDES) permit, and the Consumer Confidence Report. After all the information gathered, coordinates were added visually using satellite imagery in ArcGIS, Google Earth, and Google Maps to proceed to the ground-truthing step. The actual (ground-truthed) coordinates identified as its location along the river reach or water bodies were updated as an input geospatial dataset for the DRINCS model. The attribute data of the two joined layers were compared to ensure matching the name of effluent-receiving rivers or surface water sources. This process for both WWTP and DWTP locations are manual and time-consuming. Thus, when the DRINCS was created, the model was limited to include only 2,056 surface water intakes at 1,210 large DWTPs (serving > 10,000 people) and only 20% of WWTP locations were ground-truthed (Rice & Westerhoff, 2015). Although 64% of the U.S population has been served by the large DWTPs, the number of small DWTPs is double the number of large systems (N = 3,984) (USEPA, 2019b). The need for expanding the DRINCS model is central to this effort to include the prediction of all surface water intakes at small DWTPs (serving ≤ 10,000 people) in a systematic analysis of de facto reuse present in source surface water for large versus small DWTPs across the U.S. The modeled results can help to understand the risks influencing communities of different population sizes in the U.S.

The spatially explicit DRINCS model allows the development of new functionality based on ArcGIS application and automation of workflows (Zandbergen, 2015). ArcGIS provides users multiple existing geoprocessing tools for processing geographic and related data. Users can build custom tools in the support of geoprocessing or use a comprehensive suite of geoprocessing tools to perform spatial analysis or manage GIS data in an automated way (Zandbergen, 2015). Python is an interpreted language that works directly with available functions in ArcGIS as it was embedded in many tools in the ArcGIS programming for Desktop. Furthermore, users can turn their

Python script into a script tool to access the existing functionality of ArcGIS and extend that functionality (Zandbergen, 2015). A script tool allows users to perform the geoprocessing operations without having to work directly with the Python code as it owns a dialog box that provides an easy-to-use interface for selecting input data before executing the script. Script tools can be easily added or integrated into a model tool in ModelBuilder to perform an automated workflow in ArcGIS (Zandbergen, 2015).

This chapter overviews recent developments of the DRINCS model regarding the expanding number of water systems, updating with the most recent database and developing a new model tool based on ArcGIS application.

Objective 1: Expand the DRINCS model to include all small DWTPs (serving ≤ 10,000 people) and update the primary database of the DRINCS

Objective 2: Develop a model tool of proximity analysis between upstream WWTP and DWTPs at a watershed level

Expanding the DRINCS model

The expanded DRINCS obtained DWTP data from the SDWIS/Federal calendar year 2019 which included 6,045 surface water intakes at 3,984 small DWTPs (serving ≤ 10,000 people) for all 48 contiguous states in the US, District Columbia, and the tribal regions (USEPA, 2019b). This source of the database is also used to update the basic water system information and DWTP locations for the existing data of large DWTPs (serving > 10,000 people). Nationally, the DRINCS includes an updated data of 9,702 surface water intakes at all sized DWTPs (N = 5,575) and these surface water intakes have been ground-truthed completely (Figure 4.1). A public water system is classified as an active or inactive system. A system is active if it produces drinking water regularly or seasonal if its operation is expected to resume within the year. Inactive public water systems are the systems that already went out of business or were merged into other PWSs (USEPA, 2019a). It is noted that DWTP data from the EPA SDWIS/Federal are not real-time data. Data from each calendar quarter become available in the SDWIS system after the end of the following quarter. The date of the latest SDWIS data is also available in the SDWIS database (USEPA, 2019a). In some states, more recent data on DWTPs can be obtained from their websites, for example, the Consumer Confidence Report, the Environmental Department of the state, or the Drinking Water Watch website of the state.

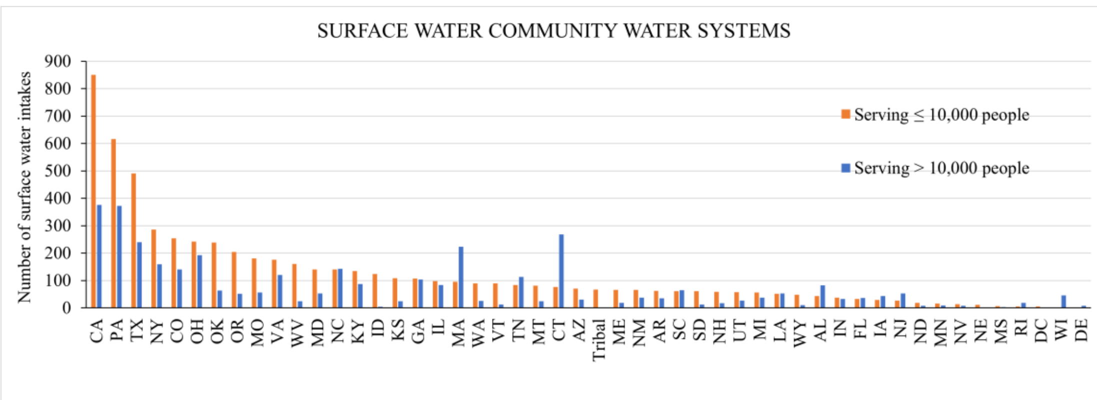

Figure 4.1 Number of all Surface Water Intakes by DWTPs at each State in the DRINCS Model Across the United States (N = 9,702)

The most recent WWTP database is the Clean Watersheds Needs Survey (CWNS) 2012, released in May 2016 (USEPA, 2016). The CWNS 2012 did not report the data for some states, such as South Carolina, American Samoa, and N. Mariana Islands as they did not participate in the survey. As South Carolina is included in the DRINCS, the missing information for this state in CWNS 2012 can be derived from the data in CWNS 2008. Some data gaps in CWNS are unique to each facility such as design capacity, WWTP locations, and level of treatment. These were filled by the values in the previous year's report. Thus, in the DRINCS, the compilation of the CWNS dataset was completed with 16,161 WWTPs through the CWNS 2004, 2008, and 2012. Previously only 20% of WWTP outfall locations were ground-truthed (Rice et al., 2015). The research in this dissertation has completed the ground-truthing work for all WWTP wastewater outfalls (N = 16,161) (Figure 4.2).

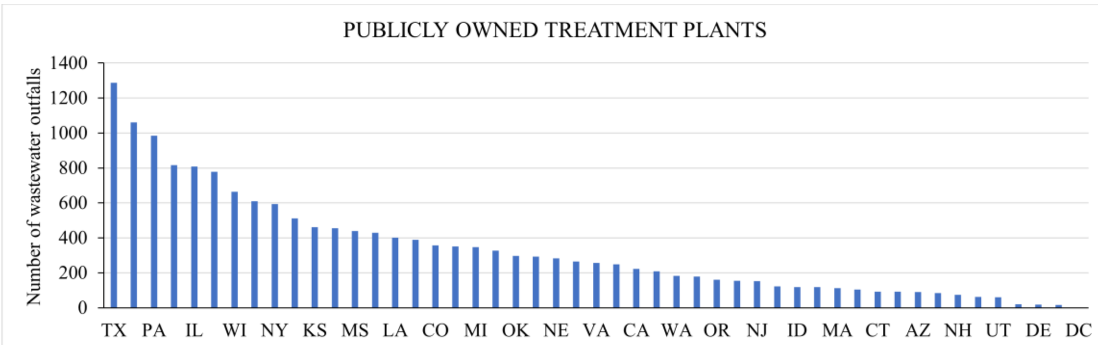

Figure 4.2 Number of Wastewater Outfalls into Surface Water from Publicly Owned Treatment Plants in the Compilation Dataset of CWNS 2004, 2008, and 2012 (N = 16,161)

The National Hydrography Dataset Plus (NHDPlus) provides integrated and application-ready vector data and attribute tables (NHDPlus, 2012). The NHD Plus based on the medium resolution (1:100,000 scale) includes water bodies, watershed boundary, stream networks with more than three million features. The NHD Plus Version 1 was created in 2006 and it was modified as the NHD Plus Version 2 (NHD PlusV2) released in 2012 (NHDPlus, 2012). The NHD PlusV2 Attributes have been updated in 2017 while the feature database maintains the same. This updated attribute used in the DRINCS model includes meaning annual streamflow, mean annual velocities, and Strahler stream order. One of the major changes in the NHD Attribute is the addition of new calculated values, for example, mean monthly flow estimates and velocities and travel time for rivers and lakes for all stream network. The Enhanced Runoff Method (EROM) was used to calculate these values and valid for the 1971 to 2000 time period. The DRINCS model took advantage of this updated NHDPlusV2 Attribute for further analysis of the DRINCS results (NHDPlus, 2012). For example, the occurrence of de facto reuse at different Strahler stream orders and calculation of the proximity between WWTPs and DWTPs was determined based on velocities and travel time. Furthermore, the processing units for vector data of the NHD PlusV1 were referred to as the "Hydrologic Region" or HUC in the United States. These processing units in the NHD PlusV2 were updated with the "Vector Processing Unit" (VPU) for vector data (NHDPlus, 2012). The change from HUC to VPU for vector data reflects the change in the number of units to process which is 18 and 12, respectively for the contiguous United States. In figure 4.3, some VPUs in the NHD PlusV2 are different from HUCs in the NHD PlusV1 while others are the same. Some HUCs are grouped into a VPU, for example, the Mississippi VPU is a group of multiple HUCs (05, 06, 07, 08, 10L, 10U, and 11) to consider the watershed connections among these HUCs (NHDPlus, 2012). The proximity model tool of the DRINCS model was developed based on the VPUs for vector data and the flow direction attribute data in the NHD PlusV2.

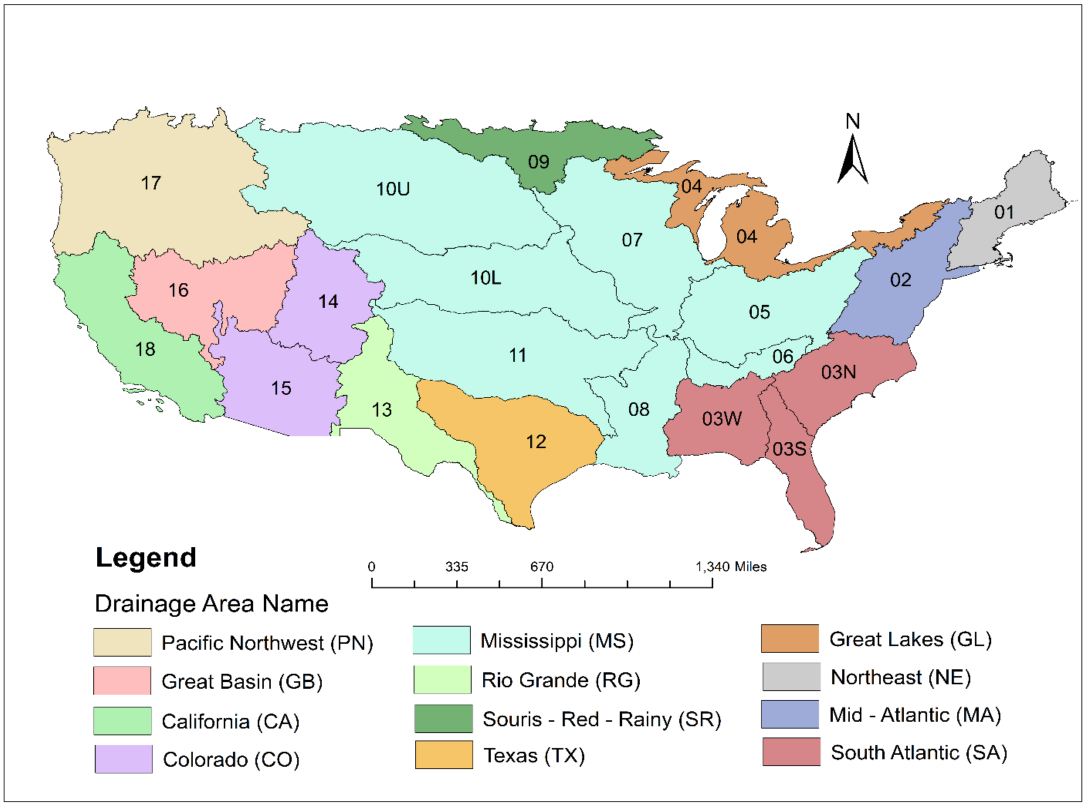

Figure 4.3 Processing Units of Vector Data in NHD PlusV2

In conclusion, nationally, the DRINCS 2.0 has been updated and expanded to contain a completely ground-truthed dataset for all sized DWTP intakes (N = 9,702), WWTPs (N = 16,161), and updated values for mean annual flow for the U.S rivers (N ~ 3 million) to quantify the de facto reuse present at surface water sources. The study in Chapter 6 analyzed all active DWTP intakes (N = 6,826) at 3,947 public water systems across the United States. A new proximity model tool (in this chapter) was developed based on the updated NHD PlusV2 Attributes for VPUs, velocities, and travel time. The DRINCS 2.0 modeled results are further discussed in Chapter 6 which is in preparation for submission to a peer-review journal.

Development of a Proximity Model Tool

The need to understand the potential for attenuation of contaminants to occur between the locations where the treated effluent enters surface water and drinking water intakes facilitated the development of a new tool in the DRINCS model to automate and determine the proximity analysis associated with de facto reuse.

Before creating an automated tool, an ArcGIS based conceptual model of the proximity analysis has been developed in a publication in Chapter 3 and continued to pilot for a case study in a publication in Chapter 5. At the time, the calculation of the proximity was quite time-consuming, and no automated tools were available. This section describes how an automated Proximity model tool was developed based on ModelBuilder for ArcGIS to identify the upstream WWTPs (number and accumulated wastewater) and calculate the shortest stream path or travel time between WWTPs and DWTP. The output of the proximity model tool was used to determine the proximity indicator as a secondary index of the relative risk of having CECs of wastewater origin in DWTP source supplies. Figure 4.4 describes the processing steps of the script tools integrated with the Proximity model tool.

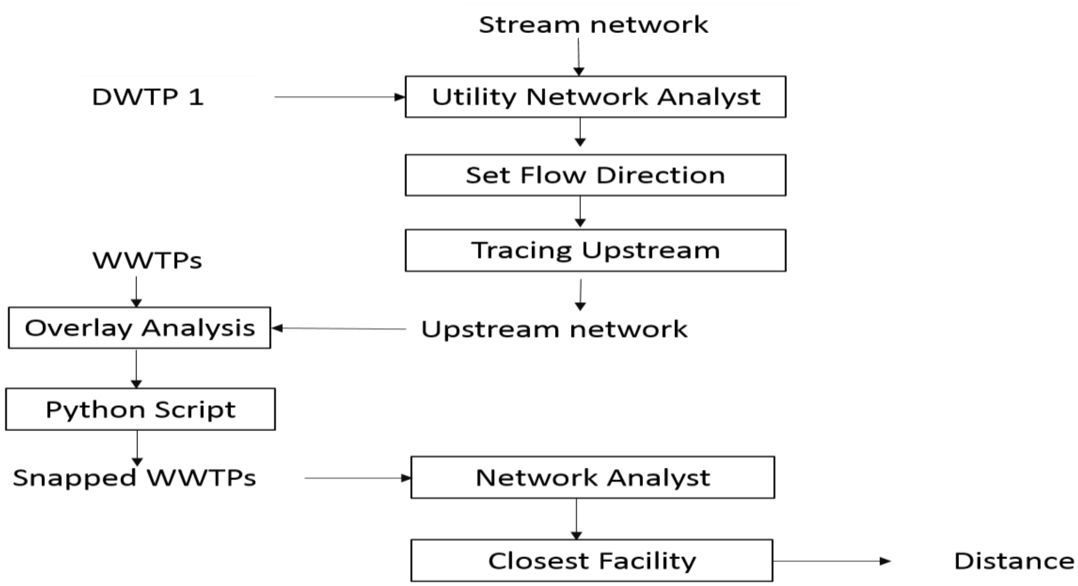

Figure 4.4 A Conceptual Model of the Proximity Model Tool

The Proximity model tool built on a ModelBuilder implements two main script tools and the other two scripts for pre-processing of the data. When a tool is executed, a script is run to carry out the geoprocessing operations. The interface of the tool dialog box provides an easy-to-use interface for specifying the input parameters, output datasets, and other control parameters before it is executed (Figure 4.5).

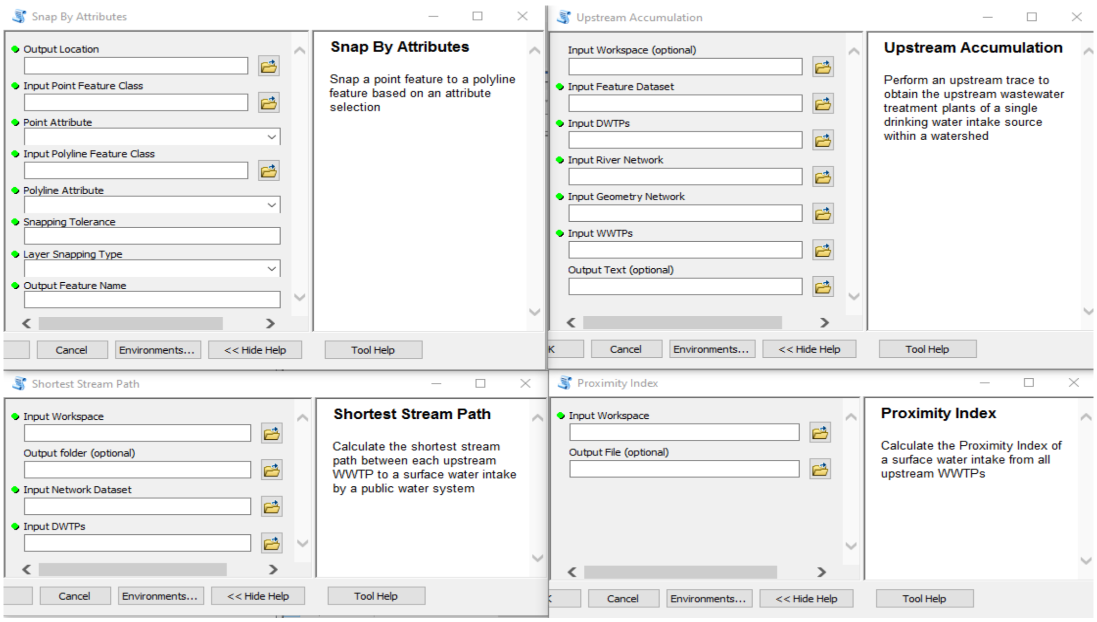

Figure 4.5 Screenshot of a Tool Dialog Box for a Script Tool in ArcGIS

The Proximity model tool is a programmed ArcGIS tool to map and determine the proximity between the WWTPs discharging into the receiving rivers and DWTP intake. The new model tool is created within ModelBuilder to integrate four script tools into the model which are, Snap by Attribute, Upstream Accumulation, Shortest Stream Path, and Proximity Index (Figure 4.6). Excluding the proximity index tool, the conceptual model was interpreted in Python language to work directly with the existing functions available in ArcGIS to create three out of four script tools. Python is integrated within the ArcGIS 10.

This study used Python 2.7, ArcMap™ 10.4.1, and the Network Analysis Extension. Three script tools were created from original Python scripts to perform the extended functionality of the ArcGIS toolbox in an automation way. Python makes script tools work like a geoprocessing tool in ArcGIS. ArcPy is a Python site package that provides a dynamic way to perform geoprocessing without opening an ArcGIS. This geoprocessing is no limited including geographic data analysis, data conversion, data management, and map automation with Python.

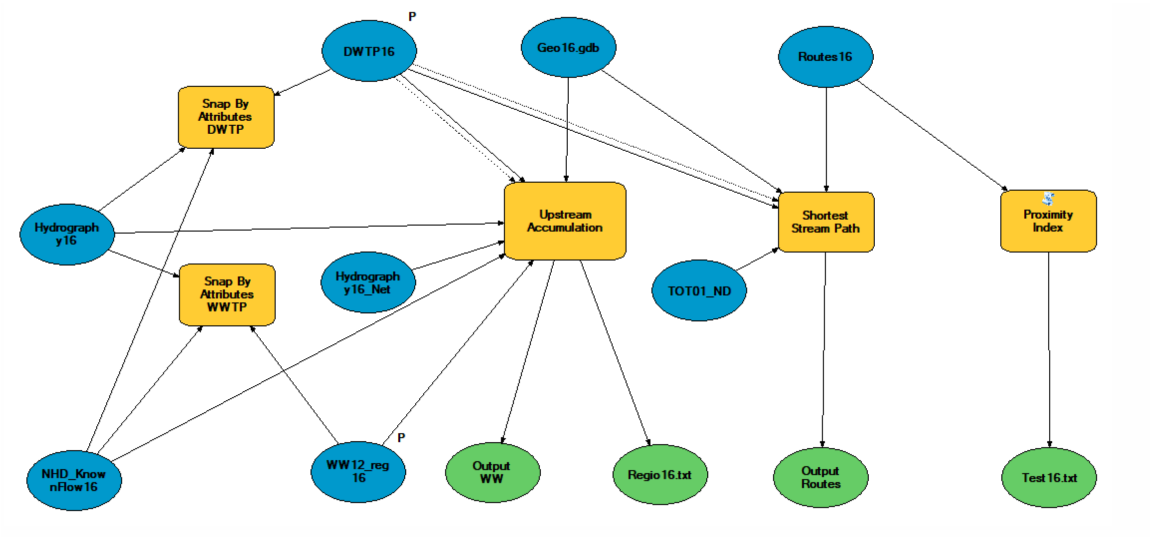

Figure 4.6 The ModelBuilder Interface of the Proximity Model Tool

ArcGIS provides ModelBuilder as a tool to create a model tool by using a visual programming language. A set of script tools can be integrated into a ModelBuilder for faster and user-friendly execution, in which the output of one tool becomes the input for another tool.

Snap by Attribute script tool. This is a script tool to pre-process the feature class before running in the model. The Snap by Attribute tool used the edited snap tool in the ArcGIS toolbox to move a point feature (as the location of wastewater outfall or a surface water intake) to coincide exactly with the vertices, edges, or endpoints to a target polyline feature (as the river segment). The unique ID information for each river segment obtained from the previous step of ground-truthing work was compared to ensure the point was snapped on the matched river. The output was a new point feature class with the snapped coordinates in the attribute table, and then was used as the input feature for the Upstream

Accumulation tool in a geometric network (Booth & Mitchell, 2001).

Upstream Accumulation script tool. A geodatabase feature class was first created to serve as the data sources to define the geometric network. Feature class of surface water intakes, wastewater outfalls, and river networks were imported into the feature dataset. Under the feature dataset, a geometric network was built based on the feature class of the river network and set flow direction from an attribute table by using an ArcGIS toolbox. Edge and junctions were considered as lines and points in the network and the flow was transferred from one edge to another through junctions. A geometric network modeled the flow of treated wastewater along the rivers (edges) that were connected by the river (junctions) with the flow direction for each river segment from an attribute table and traced all the features upstream. The model tool is executed at a VPU watershed scale. The output result was a polyline of upstream river segments and a point feature of upstream wastewater outfalls. The attribute tables provided information on accumulated wastewater, the number of upstream wastewater outfalls, and the number of upstream river segments for each single drinking water intake (Zeiler, 1999). The output feature class from the Geometric Network script tool was used as the input parameter for the *Shortest Stream Path script tool*. The Network Analysis used the Closest Facility analysis to calculate the shortest distance along the river path between each pair of wastewater outfall and surface water intake in the network. The Closest Facility uses a multiple-origin, multiple-destination algorithm based on the classic Dijkstra's algorithm analysis of single origin-destination to find the shortest path between a pair of origin and destination location. Either feature class of upstream WWTPs and class of surface water intake were specified as origin and destination as the Network Analysis does not consider the flow direction. Depending on the impedance attribute of the network as length or travel time, the output of the tool was the calculated shortest path or travel time among these two locations stored in the attribute of a polyline feature (route) in the network (Dijkstra, 1959).

The final step was using the Proximity Index tool to perform the calculation of proximity indicator (PI) for all upstream WWTPs to a DWTP based on Equation 2, Equation 3, and Equation 4 shown in Chapter 3. One surface water intake was predicted to be impacted by multiple upstream WWTPs (up to 1,000 facilities for a case study in the Trinity River basin). The proximity distributions of WWTPs upstream to DWTPs was shown in a study for Texas (Chapter 5). Larger PI values suggest larger WWTP discharges located closer to DWTP intakes and could indicate a larger potential wastewater impact. The calculated index was further discussed in Chapter 5 of this dissertation. The proximal model tool can also provide information on the travel time of CECs in rivers which can facilitate the understanding of the fate and transport Development of the proximity model tool in ArcGIS allows an automation process of routine work, better performance with increased efficiency and productivity, and make more informed decisions and consultants in a fraction of time required.

Advances in the development of a new model tool within ArcGIS are the flexible ways of performing a geoprocessing analysis with Python. A script can be run as a stand-alone script without ArcGIS, or a script tool used a toolbox interface to specify the parameters without knowing the Python code. Stand-alone scripts can be scheduled to run at a specific time without user intervention. A series of script tools can be integrated into a model tool in a ModelBuilder to execute an automation workflow. The advanced

DRINCS model can help to integrate a wide range of disparate data sources to be combined and analyzed to support a decision-making process. When a model is a tool in an ArcGIS toolbox, it is possible and easier to save the model for future use or to share with others (Zandbergen, 2015).

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CHAPTER 5

DRINKING WATER VULNERABILITY IN LESS-POPULATED COMMUNITIES IN TEXAS TO WASTEWATER-DERIVED CONTAMINANTS

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The author's contribution is to derive the computational model, analyze the data, and write the manuscript.

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Abstract

De facto potable reuse (DFR) occurs when treated wastewater is discharged upstream of drinking water treatment plants (DWTPs) and can lead to contaminants of emerging concern (CECs) occurring in potable water. Our prior research focusing on larger communities that each serve >10,000 people across the USA indicates elevated de facto reuse occurs in Texas, so we added to our model DWTPs serving smaller communities to understand their vulnerability to CECs. Here, we show that two-thirds of all surface water intakes in Texas were impacted by de facto reuse (DFR) at levels exceeding 90% during even mild droughts, and under average streamflow, DFR levels range between 1% to 20%. DWTPs serving lower population communities (<10,000 people) have higher DFR levels, and fewer than 2% of these communities have advanced

technologies (e.g., ozone, activated carbon) at DWTPs to remove CECs. Efforts to improve water quality in these less populated communities are an important priority. The model approach and results can be used to identify prioritization for monitoring and treatment of CECs, including in underserved communities that normally lack knowledge of their impacts from de facto reuse occurring within their watersheds.

Keywords: *drinking water, wastewater, reuse, treatment, exposure*

Introduction

Wastewater discharges into the natural environment can deteriorate surface water. In the United States of America (USA), the Clean Water Act regulates municipal wastewater discharges to keep the nation's surface waters quality fishable and swimmable. The US Environmental Protection Agency (USEPA) National Pollutant Discharge Elimination System (NPDES) regulates point source discharge of wastewater to surface waters, but it rarely considers impacts on downstream drinking water treatment plants (DWTPs). Studies have detected contaminants of emerging concern (CECs), including pharmaceuticals, personal care products, and industrial chemicals, originating from wastewater in DWTPs downstream of wastewater treatment plants (WWTPs) (Benotti, Stanford, & Snyder, 2010; Benotti et al., 2009; Bieber, Snyder, Dagnino, Rauch-Williams, & Drewes, 2018; Nguyen et al., 2018; Schultz et al., 2010). A previous study on the 50 very large WWTPs (between 15 and 660 million gallons per days (MGD)) across the US reported 6,000 MGD (263 m^3/sec) discharging to surface waters and measured 56 active pharmaceutical ingredients in effluent samples (Kostich, Batt, & Lazorchak, 2014). Additionally, some CECs lead to N-Nitrosodimethylamine disinfection by-product formation in drinking waters after chlorination (Hanigan et al., 2015; Rice, Via, & Westerhoff, 2015; Rule, Ebbett, & Vikesland, 2005). Furthermore, the public perception of CECs is unfavorable, despite evidence of their minimal human health risk because of the low exposure potentials in drinking water (Anumol, Clarke, Merel, & Snyder, 2015; Bruce, Pleus, & Snyder, 2010; Rice, Wutich, White, & Westerhoff, 2016; Stanford, Snyder, Trenholm, Holady, & Vanderford, 2010).

De facto reuse occurs when a municipality withdraws water from a river or reservoir that includes treated wastewater discharged from upstream WWTPs (Reuse, 2012; Rice, Wutich, & Westerhoff, 2013). The previously developed De Facto Reuse Incidence Nations Consumable Supply (DRINCS) model by Rice and Westerhoff (2015) analyzed treated municipal wastewater discharges from WWTPs and included combined sewer systems, although it does not consider combined sewer overflows or wet weather by-passes. The DRINCS model has been used and validated through field sampling in several case studies (Barber et al., 2019; Nguyen et al., 2018; Rice et al., 2015; Rice et al., 2013). Our prior DRINCS study concluded that >50% of DWTPs in the US serving 10,000 or more people with treated surface water have at least one WWTP discharge upstream of the drinking water intake (Rice & Westerhoff, 2015). While the frequency of de facto reuse is high, its magnitude is relatively low under average streamflow conditions. That previous DRINCS study, that considered only DWTPs serving 10,000 people or more, found among the highest de facto reuse occurs in the Texas Gulf region (USGS Hydrologic Region 12) with de facto reuse occurring at 90% of the DWTP intakes. Other studies also indicate high levels of wastewater in surface waters in Texas (Brooks, Riley, & Taylor, 2006; Slye et al., 2011). Therefore, this paper focuses on the state of Texas (USA) and DWTPs that contrasts larger (>10,000 people) to smaller (<

10,000 people) sized communities.

CECs undergo biogeochemical transformations (e.g., hydrolysis, oxidation, hydroxylation, conjugation, cleavage, de-alkylation, methylation, and demethylation) in surface waters, and the transformations are impacted by stream geometry and travel times (Challis, Hanson, Friesen, & Wong, 2014; Chen, Nam, Westerhoff, Krasner, & Amy, 2009; Radke, Ulrich, Wurm, & Kunkel, 2010). Transformation products are often more polar, less bio-accumulative, and can be less toxic than parent compounds in the aqueous environment (Boxall, Sinclair, Fenner, Kolpin, & Maund, 2004; Rice et al., 2015; Rice et al., 2013). However, some derivative compounds can be more persistent and may have adverse human health effects (Cwiertny, Snyder, Schlenk, & Kolodziej, 2014). CEC removal at DWTPs depends on raw water quality, the chemical structure of target CECs, and specific unit processes in place (Liu, Kanjo, & Mizutani, 2009; Westerhoff, Yoon, Snyder, & Wert, 2005). Prior DRINCS modeling and nearly all field campaigns to quantify de facto reuse has focused on DWTPs serving larger populations (e.g., > 10,000 people), thereby potentially overlooking impacts from de facto reuse on DWTPs serving smaller (or potentially underserved) communities. The value of the data science approach behind DRINCS can allow screening or prioritization for CEC monitoring or treatment, after the inclusion of DWTPs serving smaller-sized communities (< 10,000 people) are included in the models.

In this study, we expanded the DRINCS model from only 156 DWTP intakes serving 10,000 or more people to include all DWTPs in Texas (US) by locating, ground-truthing, and adding 244 DWTP intakes serving 10,000 or fewer people (Rice & Westerhoff, 2015). De facto wastewater reuse was modeled under average and variable streamflow conditions. DRINCS only includes treated and discharged wastewater effluent, but not contributions from stormwater discharges or non-point sources (e.g., septic systems, surface runoff). Using a Dijkstra's algorithm by Dijkstra (1959), proximal distances between WWTP discharges and DWTP intakes, and their frequency distribution when multiple WWTPs were located upstream, were incorporated for the first time into DRINCS. Using information about the specific unit processes installed at each DWTP, we evaluated the capability of the DWTPs to remove CECs, should they be impacted by upstream WWTP discharges (i.e., de facto reuse). This information was then used to discuss social equity issues and the need to increase CEC monitoring in less populated, often rural, communities.

Result and Discussion

DFR occurrence and magnitude under mean annual streamflows. Figure 5.1 shows the spatially distributed levels of de facto reuse (DFR) under mean streamflow conditions for all DWTPs with surface water intakes within Texas. Two-thirds of the DWTP intakes (422 out of 595) were impacted by the potential presence of wastewater, as defined as having at least one upstream wastewater discharge. This includes 222 intakes at 182 DWTPs that serve populations of ≤10,000 (Table 5.1). DWTP intakes impacted by at least one upstream WWTP discharge included intakes located on lakes and reservoirs (n = 225), streams or creeks (n = 108), or canals (n = 89). While the frequency of de facto reuse is high (~67%), roughly 60% of impacted DWTP intakes have <5% DFR under mean annual streamflows; 5% DFR equates to 5% of the water at a DWTP intake potentially being of wastewater origin based upon Equation 1. However, DFR was higher in southwestern Texas with most having >20% DFR under annual mean streamflows. 34 surface water intakes by DWTPs supplied by the Rio Grande in the southwestern of Texas (Strahler stream order = 8) have high DFR (>20%). There were 173 surface water intakes by 97 DWTPs in Texas not impacted by upstream WWTP discharges, and 61 of these serve 10,000 or fewer people.

Strahler stream orders play an important role in DFR magnitude at drinking water sources (Rice & Westerhoff, 2015). Figure 5.2 shows that the highest DFR levels were in the smallest and largest rivers in Texas, or lower to higher Strahler stream orders. DFR varies substantially among DWTPs located on different stream orders. First-order streams are smaller and therefore rely more on WWTP discharges to maintain even mean annual streamflows. Hence, smaller streams are more likely to contain CECs throughout the year. Most DWTPs on 2nd through 5th order streams had DFR below 5%. In contrast to national trends where DWTP intakes on higher stream orders have lower DFR (Rice & Westerhoff, 2015, 2017), presumably due to natural runoff diluting wastewater, streams of 6th, 7th, and 8th order in Texas show higher DFR. This illustrates how geographical location within the arid southwestern US can impact DFR perhaps more than general stream order classification on a national basis. Nearly all DWTPs on the higher stream orders are in Texas were impacted by at least one upstream wastewater discharge.

Effects of variable streamflow on DFR magnitude. Reduced streamflow during drought may increase de facto reuse. However, unlike the mandated requirement to have streamflow data or predictions at the WWTP discharge locations to calculate dilution factors, there are rarely in-stream gauging stations or long-term streamflow datasets available at DWTP intake locations. Lack of long-term (>30 years) data limits the ability to perform statistical analysis of drought or flood impacts on DFR. With the use of the

USGS stream gauge database within the NHD plus suite, 22 of the 595 DWTP intakes had adequate long-term (>30 years) historical streamflow data, and DFR trends as a function of increasing streamflow were assessed. Figure 5.3 illustrates DFR for DWTPs as a function of both Strahler stream order and historical streamflow. 15 of the 25 sites have >10% DRF at the 50th percentile flow. At the 7Q2 condition (~10th percentile streamflow), treated wastewater made up ~100% of the water supply for 14 of 25 DWTP intakes. During seasonal low flow or drought periods, which is the design condition for WWTP effluents, there is a high occurrence of DFR (and associated CECs) at downstream drinking water intakes.

Proximity distribution of WWTPs upstream to DWTPs within the Trinity River Basin. Figure 5.4 shows the location of 151 WWTPs discharges upstream of 10 surface water DWTP intakes in the Trinity River Basin. Located at the upstream end of the lake, DWTP 10 withdrew water from Lake Lewisville, while DWTP 04 and DWTP 05 also used Lake Livingston as a drinking water source. Other DWTPs withdraw water from tributaries (Elm Fork Trinity River) or mainstream of the Trinity River.

Twenty-one WWTPs influenced the most up-river drinking water facility (DWTP 10), whereas 151 WWTP discharges were upstream of DWTP 01. Because WWTPs discharge into tributaries and the mainstream of the Trinity River, linear addition of treated wastewater does not occur. Instead, there is a distribution of distances from different tributaries that affect an individual DWTP. Figure 5.5 and Table SI.3 present cumulative distributions for the number of WWTPs located at different distances upstream from each of the 10 DWTPs. Figure SI.4 and Table SI.2 show cumulative wastewater discharges, instead of the number of facilities, using a similar x-axis. There are no DWTPs within this watershed with a WWTP located fewer than 16 km upstream (10 miles), except two facilities (DWTP 04 and 10) are located on lakes that receive WWTP discharge. Lakes can have complex stratification and mixing patterns and would necessitate site-specific hydrologic modeling to understand precise levels of DFR. However, DRINCS helps identify such site-specific needs. Eight DWTPs have WWTPs located 16–40 km upstream (10–25 miles), and most of the WWTP discharges are located 160–500 km (100–300 miles) upstream of DWTP intakes. Figure SI.4 illustrates cumulative wastewater upstream for each of the ten DWTPs in the Trinity River basin. The wastewater volume varied from less than 10 MGD (within <16 km) to nearly 1,400 MGD (>1,600 km).

Travel time of CECs in rivers can reduce their concentrations through biogeochemical transformations. Travel time can be calculated by dividing the distance by streamflow velocity. However, velocities depend on volumetric flowrate, drainage area, rainfall intensity-frequency-duration relationships, gradient or slope of the riverbed, and cross-sectional area of the channel. For lakes and reservoirs, NHD Plus identified streamlines were used to calculate travel times and then CEC attenuation; more detailed lake mixing models could be pursued in the future that includes lake stratification or mixing and hydraulic residence times. High velocities often occur during the flood or other high streamflow events, where greater wastewater dilution occurs and thus is probably less important for CECs than lower flow periods (Benotti et al., 2009). Typical stream velocities are 0.15–0.6 m/sec (0.5–2 ft/sec), but they can be slower under low streamflow conditions. Travel time estimates are shown in Figure SI.5. Streamflow of 0.3 m/sec would result in travel times of 0.6, 1.5, 6.2, and 19 days for 16, 40, 160, and 500 km, respectively. CEC half-lives in surface waters can range from hours (e.g., photo-labile) to months (e.g., artificial sweeteners), depending upon their reactivity. For seven CECs commonly used as surrogates (Dickenson, Snyder, Sedlak, & Drewes, 2011), we applied EPI Suite™ and fate model LEV3EPI™ to estimate half-lives (Muñoz et al., 2008). Table SI.4 shows the degradation rates of the seven CECs in water. Of the compounds studied, ibuprofen had the shortest half-life in water (15 days); diclofenac, meprobamate, gemfibrozil, and sulfamethoxazole SMX were next at 37.5 days; and TCPP and TCEP had the longest half-life (60 days). CEC attenuation with distance was estimated using pseudo-first order degradation kinetics (Morrall et al., 2004):

$$C(t) = C_i * e^{-kt} \qquad \text{Equation 2}$$

Where: $C(t)$ = analyte concentration at time t

C_i = initial analyte concentration

k = first-order transformation rate (1/day) and $\mathrm{k} = \frac{\ln 2}{t_{1/2}}$

$t_{1/2}$ = half-life of CEC in water (days)

t = travel time (days), calculated as distance divided by streamflow velocity

Typical streamflow velocities range between 0.05 and 0.5 m/sec, resulting in travel times of 6 to 60 days for a proximal distance of 250 km. Figure SI.6 shows as a function of the distance the degradation of several CECs commonly used as WWTP surrogates (meprobamate, ibuprofen, gemfibrozil, diclofenac, sulfamethoxazole, (Tris(1-chloro-2-propyl) (TCPP) and Tris(2-chloroethyl) (TCEP) phosphates) for larger CEC lists that may number in the hundreds of compounds (Barber et al., 2019; Dickenson et al., 2011; Glassmeyer et al., 2017; Nguyen et al., 2018). For a 0.1 m/sec streamflow, roughly 50% of the ibuprofen degraded within 100 km, whereas 50% degradation of diclofenac, meprobamate, gemfibrozil, or sulfamethoxazole may not be reached until 300 km. Even longer distances (600 km) may be required for similar degradation of TCPP or TCEP.

Streamflow variation impacts levels of CECs at downstream DWTP intakes in two ways: 1) lower streamflow proportionately increases CEC concentrations in rivers just below WWTP discharges (i.e., less dilution), but 2) lower streamflow proportionately lengthens hydraulic travel times that allow for more CEC attenuation via biogeochemical transformations. For the ten DWTP intakes considered in Figure 6.5 where CEC transformations similar to that predicted over 300 km may occur (Figure SI.5 and SI.6), nine DWTPs had between 20 to 30 upstream WWTP dischargers within 161 to 483 km and DWTP#7 had 65 WWTP discharges within that distance. With the distances between DWTPs downstream from multiple WWTP discharges ranging from <16 to >800 km (Figure 5.5), some CECs will probably be largely removed by natural attenuation while the concentration of more refractory CECs (e.g., TCEP, TCPP) would likely be relatively unchanged at the downstream DWTP intake.

Unit processes at DWTPs Impacted by De Facto Reuse. Water treatment plants can build and operate advanced unit processes capable of removing CECs from the intake water, in addition to conventional unit processes required to meet existing regulatory compliance. However, CECs are by their very nature "emerging" and not currently regulated. Therefore, few DWTPs are required to install advanced unit processes, unless for secondary benefits (e.g., reduction in algae-derived tastes and odors) or necessity to meet disinfection and disinfection by-product rules established by the USEPA. This section uses data from the State of Texas on the type of unit processes installed at

DWTPs to explore which facilities, as a function of their size and impact by de facto reuse, employ advanced unit processes that would be able to remove CECs. Figure 5.6 and Table SI.5 summarize the unit processes installed at all DWTPs in Texas and also for the subset of DWTPs impacted by de facto reuse. Each DWTP combines several unit processes that will achieve variable CECs removal efficiencies. 236 DWTPs impacted by wastewater in Texas disinfect using chloramines. DWTPs using free chlorine can also form chloramines if ammonia from upstream WWTPs is present. Chloramines react with some CECs to produce N-Nitrosodimethylamine (NDMA) and other probable carcinogens (Hanigan et al., 2015). Prior work shows correlations between detectable NDMA at DWTPs with DFR>0 by Rice et al. (2015), suggesting that CEC removal may be necessary. State-of-the-art unit process trains for planned, direct potable reuse include 1) reverse osmosis followed by advanced oxidation; 2) riverbank filtration; or 3) ozonation followed by biofiltration; followed by an environmental (groundwater aquifer, surface water) or engineered buffer (Gerrity et al., 2011; Gerrity, Pecson, Trussell, & Trussell, 2013; Hollender et al., 2009). However, comparable strategies currently do not exist for DWTPs with de facto reuse.

Conventional treatment processes (i.e., coagulation, sedimentation, and filtration) are used at >80% of the DWTPs in Texas. However, the conventional unit processes achieve <30% CEC removal (Westerhoff et al., 2005). Ultra- or microfiltration provides only minimal improved performance in CEC removal compared to granular media filtration. Advanced oxidation processes (e.g., ozonation or ultraviolet (UV) irradiation alone or with hydrogen peroxide (H_2O_2)) are effective in removing CECs (Dickenson et al., 2011; Heberer, 2002; Westerhoff et al., 2005). However, only 13 of 303 (5%)

DWTPs that are impacted by DFR use these unit processes; DWTPs uses ozonation alone (n=10) or with hydrogen peroxide (n=2), and ultraviolet with hydrogen peroxide (n=1). Physical removal of CECs can be achieved by sorption to activated carbon or separation using nanofiltration or reverse osmosis membranes (Kim et al., 2018; Scheurer, Storck, Brauch, & Lange, 2010; Sophia A & Lima, 2018; Wols & Hofman-Caris, 2012; Yu, Peldszus, & Huck, 2008). 43 of 303 DWTPs in Texas with DFR>0 use activated carbon (both in granular and powder one) and only 9 of those DWTPs impacted by DFR use granular activated carbon (GAC). GAC is often used at DWTPs to control algal-related taste and odors, DBP precursors, and more recently CECs. Seven DWTPs were impacted by the DFR report using reverse osmosis.

DWTP treatment disparity for low population communities impacted by de facto reuse. Many DWTPs (N=303) in Texas are impacted by at least one upstream WWTP (Figure 5.1), including 182 DWTPs serving 10,000 or fewer people, of those are 120 DWTPs serving 3,300 or fewer people. However, because the advanced DWTP unit processes are not uniformly applied at smaller and larger DWTPs, CEC exposure in treated drinking waters varies. Figure 5.7 shows the distribution of DWTP levels of treatment by population served, and whether the DWTP is impacted or not by de facto reuse. Figure 5.7 also superimposes whether or not the unit processes at the DWTP are capable of removing CECs (i.e., advanced treatment). For this analysis, we considered advanced treatment processes those with the highest potential to remove CECs: ozone alone or with hydrogen peroxide, granular activated carbon, or reverse osmosis. As summarized in Table 5.1, the majority of DWTPs serving smaller communities (< 10,000 people) did not employ advanced treatment (Figure 5.7), and the percentage not employing advanced treatment was even higher (90%) among the smallest DWTPs (serving <3,300 people). Less populous communities with smaller DWTPs often lack the financial capacity (e.g., taxation base) to fund capital investment and higher operational costs associated with advanced treatment unit processes. Needs exist to provide financial mechanisms to encourage the installation of more advanced drinking water processes at "higher-risk" DWTPs (i.e., those with higher DFR).

There are many reasons advanced technologies are not installed at facilities serving smaller communities (<3,300 or 3,300–10,000). The disparity in drinking water quality in systems serving smaller versus larger populations is evident in the number of violations across the US for existing USEPA regulations (Allaire, Wu, & Lall, 2018). For example, in Texas, Table SI.7 and SI.8 show nearly 70% of the maximum contaminant level (MCL) violations occur at systems serving fewer than 10,000 people. Figures SI.9 and SI.10 shows the most commonly reported violations are total trihalomethanes (TTHM) and five haloacetic acids (HAA5). By their very nature, CECs are "emerging" and hence are unregulated; thus, they are not part of health-based water quality violations at DWTPs (DeFelice, Leker, & MacDonald Gibson, 2017). We considered relationships between cancer mortalities and de facto reuse, but as many CECs are pharmaceuticals they do not cause cancer but rather potentially endocrine disruption or several other endpoints that have yet to be epidemiologically supported at low concentrations that occur in drinking waters (Bruce et al., 2010). One use of this paper could be to locate potential communities for inclusion in such toxicology by (Barber et al. (2019); Zhen et al. (2018))or epidemiology studies.

Implications. This study found that 303 DWTPs in Texas were impacted by at least one upstream WWTP (Figure 5.1), including 182 DWTPs serving <10,000 people with 120 of those DWTPs serving <3,300 people. Smaller communities are more commonly located on lower stream order (Strahler stream order 1^{st} to 5^{th}; Figure SI.7). Thus, more of small DWTPs are likely impacted by CECs in Texas. Using similar methodologies as applied herein, de facto reuse levels covering the same orders of magnitude as reported herein are being predicted globally (Hass, Duennbier, & Massmann, 2012; Hristovski, Pacemska-Atanasova, Olson, Markovski, & Mitev, 2016; Kapo et al., 2016; Karakurt, Schmid, Hübner, & Drewes, 2019; Loos et al., 2009; Simazaki et al., 2015; Wang, Shao, & Westerhoff, 2017). However, those studies did not focus on impacts to smaller utilities, travel times between WWTP discharges and DWTP intakes, or relate the type of treatment to the presence of CECs in DWTP intake or treated waters.

Because CECs can be transformed within surface waters, we analyzed the frequency distribution of the upstream proximal distance of wastewater treatment plants (WWTPs) discharge locations from downstream DWTP intakes. The Trinity River basin in Texas has 151 WWTPs and 45 DWTPs and was used as further modeled to understand the proximity of DWTPs from WWTPs. Most WWTPs were located 160 to 500 km upstream of DWTP intakes, where travel times between potential CEC sources and DWTP intakes range from 5 to 15 days under average streamflow. This leads to environmental exposures but allows time for in-stream biogeochemical processes to transform some CECs. This study also found that fewer than 10% of smaller sized DWTPs in Texas employ advanced technologies capable of removing CECs. Because these small communities have among the highest DFR levels, there is a need to increase resources to prioritize monitoring and installing advanced treatment in these facilities. To date, most CEC field occurrence studies at DWTPs have involved only larger-sized facilities. There is a need to involve smaller-sized DWTPs in CECs occurrence studies to understand if small systems are disproportionately impacted by de facto reuse. Analysis using DRINCS could help identify DWTPs at higher risk of de facto reuse where such studies could be most beneficial in defining the magnitude of CEC occurrence. These may also be locations where investment in public infrastructure (e.g., upgraded WWTP or advanced DWTP unit processes) may have the largest ecological or human health risk, respectively. Until such infrastructure is installed, communities predicted to have high DRF levels may be of interest to the health community as locations for assessing biomarkers or health outcomes from wastewater reuse.

Methods

Study Area and Facilities Considered. Texas is located in the south-central USA, covers 695,662 km2, and spans three national hydrologic units (Regions 11 (Arkansas White Red), 12 (Texas Gulf), and 13 (Rio Grande). Texas is the second-most populated state in the US, having about 25 million inhabitants in 2010. By 2060, the population is projected to double to 46 million people, and Texas's annual municipal water demand is predicted to increase from 4.9 million acre-feet in 2010 to 7.8 million acre-feet by 2060 (TWDB, 2017). Water availability varies in Texas, spanning from limited resources in the arid western region to being water-rich in eastern areas (Stillwell, King, Webber, Duncan, & Hardberger, 2011). Increasing populations will likely lead to greater reliance and impacts of planned and unplanned (de facto) wastewater reuse.

This study included 400 community public water systems in Texas withdrawing surface water from 595 surface water intakes (Table 5.1). Some DWTPs have more than one surface water intake. Figures SI.1 and SI.2 show locations of drinking water sources in Texas and the population served by each DWTP. These surface water DWTPs account for 2,690 million gallons per day (MGD) of design capacity (118 m^3/sec). This is augmented with a large number of groundwater-supplied facilities that combine to treat up to another 1,514 MGD (66 m^3/sec) of potable water. The groundwater facilities were not included in this study because de facto reuse is less common in groundwater systems and is not considered in the DRINCS model. A DWTP database was retrieved, and activity codes for facilities unified, from the Texas Drinking Water Watch (TDWW), Texas Water Development Board (TWDB), and Texas Commission on Environmental Quality (TCEQ). The databases include DWTP intake locations (latitude and longitude), public water system identification number (PWSID), population served, and additional data. Information on the type of installed treatment processes at DWTPs was obtained from the Texas Drinking Water Watch for the most recent two-year dataset available (2017). The PWSID also allows access to information on the unit processes at each facility. ArcGIS™ version 10.4 was used to create maps and conduct spatial analysis.

The data that support the findings of this study were aggregated from a variety of US Federal sources. WWTP data were obtained from Clean Watersheds Needs Surveys 2008 – Environmental Protection Agency (CWNS-EPA), which includes facility name, permit number (NPDES), level of treatment, design capacity, and location (longitude and latitude of wastewater outfalls to surface water). There are 1,206 WWTPs with a total design capacity of 3,213 MGD (141 m^3/sec) that discharge to surface waters; an additional 253 facilities discharge to groundwater, ocean, or evaporation ponds. Figure SI.3 shows that ~70% of the 1,206 WWTPs included in DRINCS for Texas is relatively small, with treatment capacity below 1 MGD (0.05 m^3/sec).

Predicting De Facto Reuse (DFR) Using DRINCS. The ArcGIS-based model of De Facto Reuse Incidence in our Nations Consumable Supply (DRINCS) was previously developed for all WWTPs and DWTPs serving 10,000 people or more by Rice and Westerhoff (2015) was augmented to include DWTPs serving 10,000 or fewer people from surface water sources. Precise locations of WWTP discharges and DWTP intakes were verified using the Texas Irrigation District Engineering and Assistance Program and visually ground-truthed using Google Earth. Streamflow data were obtained from the US Geological Survey - National Hydrography Dataset (NHD-USGS), and stream networks were based on the medium-resolution NHD (1:100,000 scale). Strahler Stream Order defines stream size based on the hierarchy of tributaries (Horton, 1945; Strahler, 1957), with values for the USA ranging from a low of 1st order to larger river networks that approach 9th order. Each river segment within a watershed is treated as a node, with the next segment downstream as its parent. For example, when two 1st order streams join then a 2nd order stream form. Strahler stream order can be obtained from additional calculated attributes in National Hydrography Data Plus (NHDPlus) (Pierson, Rosenbaum, McKay, & Dewald, 2008; Strahler, 1957). DRINCS was also updated by adding USGS stream gauges from within the NHDPlus suite; attribute data include average, min, max, and percentile streamflows. A key objective was to maximize the available hydrologic datasets to cover the large possible variations based upon historic streamflows data. The statistical values were calculated based on the entire record period until April 20, 2004 (the date NHD pulled the data for analysis); the starting date for each gauging station varied depending upon when it began reporting data with the earliest being November 1, 1915, and the latest was on September 27, 1997.

De facto reuse (DFR) at each DWTP withdrawing surface water was calculated as the percent of treated wastewater at a particular surface water intake, following our previously published methods and assumptions (Rice et al., 2015; Rice & Westerhoff, 2015; Rice et al., 2013):

$$\mathrm{DFR} = \frac{\sum \mathrm{WW}}{\mathrm{Q}} \mathrm{x}100\% \qquad \text{Equation 1}$$

Where Q is the streamflow at the DWTP intake location, and WW is the accumulated discharge from all upstream WWTPs, calculated by running the Python script.

In Texas, the key discharge-frequency characteristic used to evaluate the critical condition of the stream for ecological considerations is the "*annual lowest mean discharge for seven consecutive days with a 2-year recurrence interval (7Q2)*" (Texas Commission on Environmental, 2010). The 7Q2 low-flow index was calculated using an Excel-based application "Calculator for Low Flow (CALF)," which was developed by Environmental Flows Information System for Texas based on daily streamflow for 30 years of continuous USGS gauging data via Hydrologic Information System; the default period to retrieve data with CALF is from January 1, 1940, through December 31, 2009. Values for 7Q2 must be reported at WWTP discharges but are not required at DWTP intakes. Values of 7Q2 closely matched 10th percentile streamflows (from the calculations), and hence we considered low flow conditions as 10th percentile streamflows (Table SI .1– Supporting Information).

The proximity distribution between DWTPs and upstream WWTPs discharge locations was determined using digital stream networks with flow direction from the NHD Plus to build a geometric network for tracing upstream in ArcGIS. All the shapefiles were re-projected in a Texas specific projection coordinate system in ArcGIS, namely Texas Centric Mapping System/Albers Equal Area. Vector datasets of upstream segments were then used to create *ArcGIS Network Analyst* tool that calculated the stream distance between WWTPs upstream from each DWTP. Once the network was constructed, the *New Closest Facility* analysis solver was able to identify the shortest routes along with stream networks for each Facility (i.e., a single surface water DWTP intake) and Incidents (i.e., all upstream WWTP discharges) using Dijkstra's algorithm (Dijkstra, 1959). In this study, proximal distances were computed for 10 DWTPs, of which surface water intakes spanning along Trinity River in the case study in the Trinity River basin. The Trinity River was selected in part because it is one of the most populous watersheds in Texas with a total area of 17,913 square miles for the 423 mile Trinity River (TWDB) and can contain >90% wastewater effluent under low flow conditions (Fono, Kolodziej, & Sedlak, 2006).

Data Availability

The data that support the findings of this study are available from the corresponding author upon reasonable request. Sources of the electronic dataset used within the paper are summarized in Table SI.9.

Acknowledgments

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Author Contribution

Thuy T. Nguyen conducted ground-truthing and modeling and wrote the majority of the paper. Paul K. Westerhoff developed the idea and oversaw writing.

Competing Interests

The authors declare that there are no competing interests.

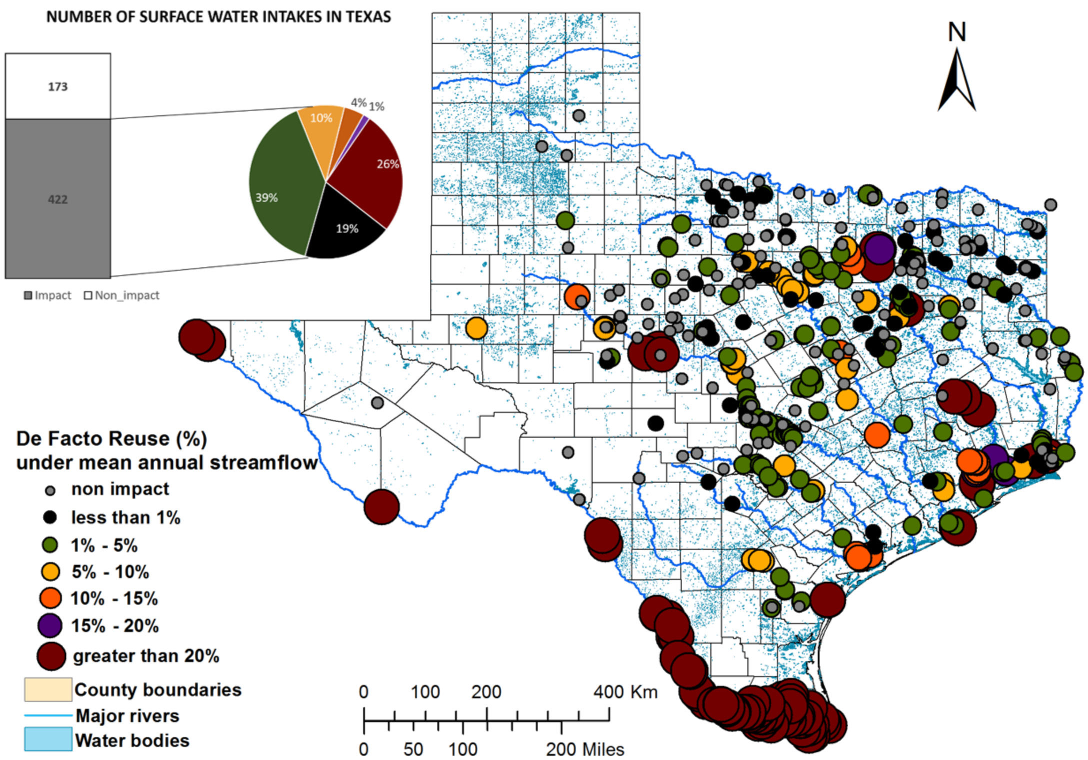


Figure 5.1 DWTPs Affected by Upstream WWTPs Discharge Under Mean Annual Stream Flows in Texas

Table 5.1 Summary of DWTPs Serving Different Community Sizes in Texas (USA)

Description	Values categorized by USEPA DWTP sizes					Totals
	Very small	Small	Medium	Large	Very Large	
Population server	≤ 500	501-3300	3301-10,000	10,001-100,000	>100,000	~ 19 million
Surface water facilities						
# Intakes (# impacted[a])	60 (39)	148 (99)	117 (84)	192 (133)	78 (67)	595 (422)
# DWTPs (# impacted[a])	49 (34)	114 (86)	82 (62)	127 (96)	28 (25)	400 (303)
DWTP with advanced Tech[b] in category size						
From All DWTPs	0%	2.6%	4.9%	7.1%	18%	5.3%
Only impacted DWTPs[a]	0%	3.5%	6.5%	7.3%	20%	4.8%

DWTPs drinking water treatment plants, USEPA US Environmental Protection Agency

[a]Indicated values are for facilities impacted by de factor reuse

[b]Advanced technology is defined as using ozonation or with hydrogen peroxide granular activated carbon, or reverse osmosis

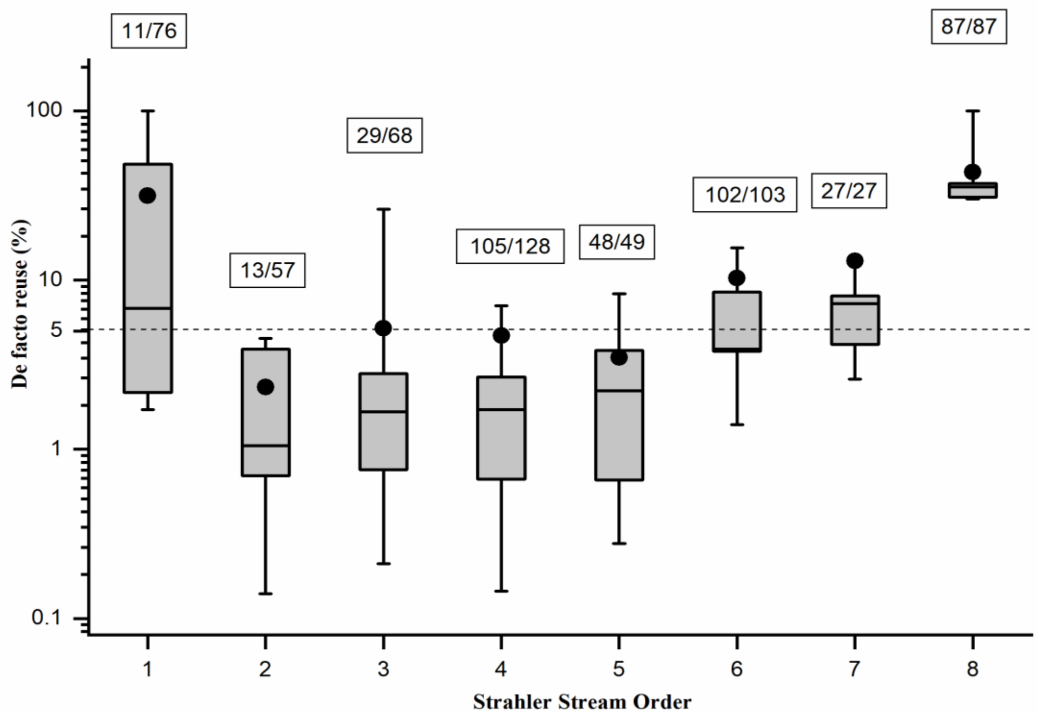


Figure 5.2 DFR Magnitude at DWTP Intakes Under Average Flow Condition in Texas (Top and bottom of box = 75th and 25th percentiles, respectively; top and bottom of whisker = 90th and 10th percentiles, respectively; line inside box = 50th percentile (median); dot (•) = average; dashed line = 5% DFR). Numbers above each bar-and-whisker diagram indicate the number of DWTP intakes with DFR>0 included in the analysis for each stream order relative to the total number of DWTPs in Texas on surface water supplies having that stream order.

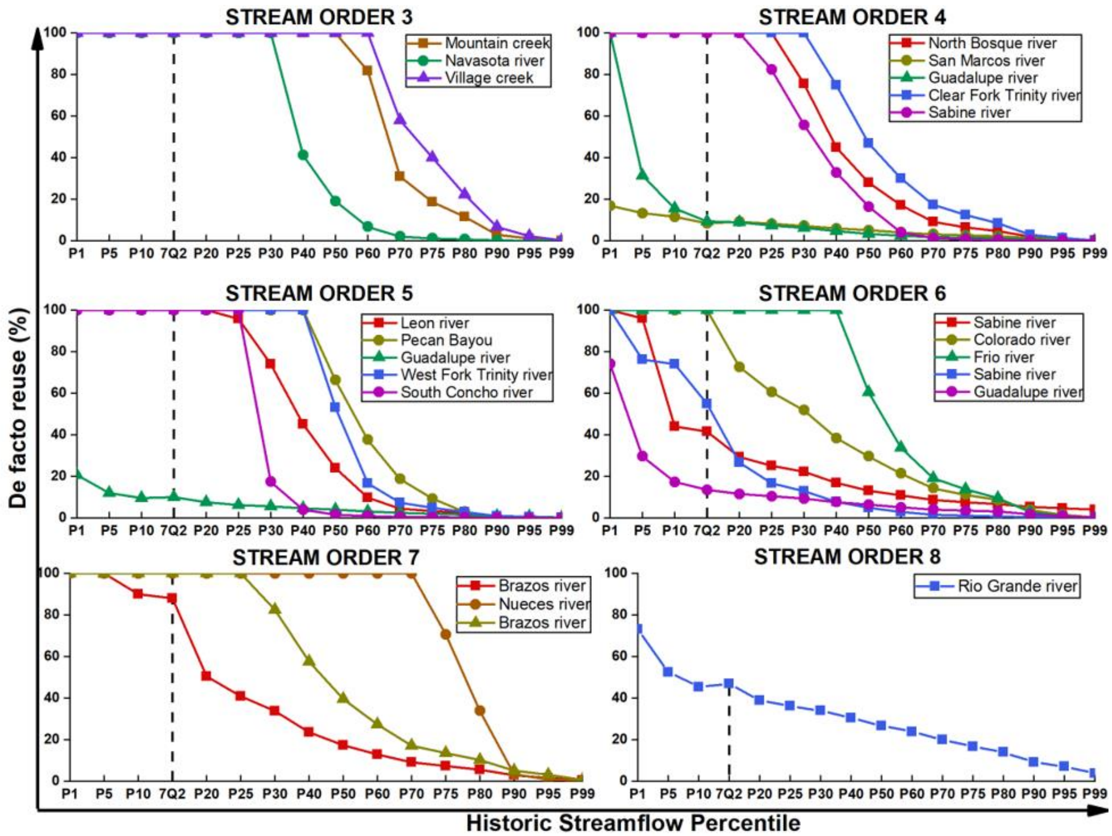


Figure 5.3 DFR variation at 22 Drinking Water Intakes in Texas Under Different Flow Conditions Across Six Different Stream Orders. Exact DWTP Locations on Each Stream are not Shown to Protect the Utility Confidentiality

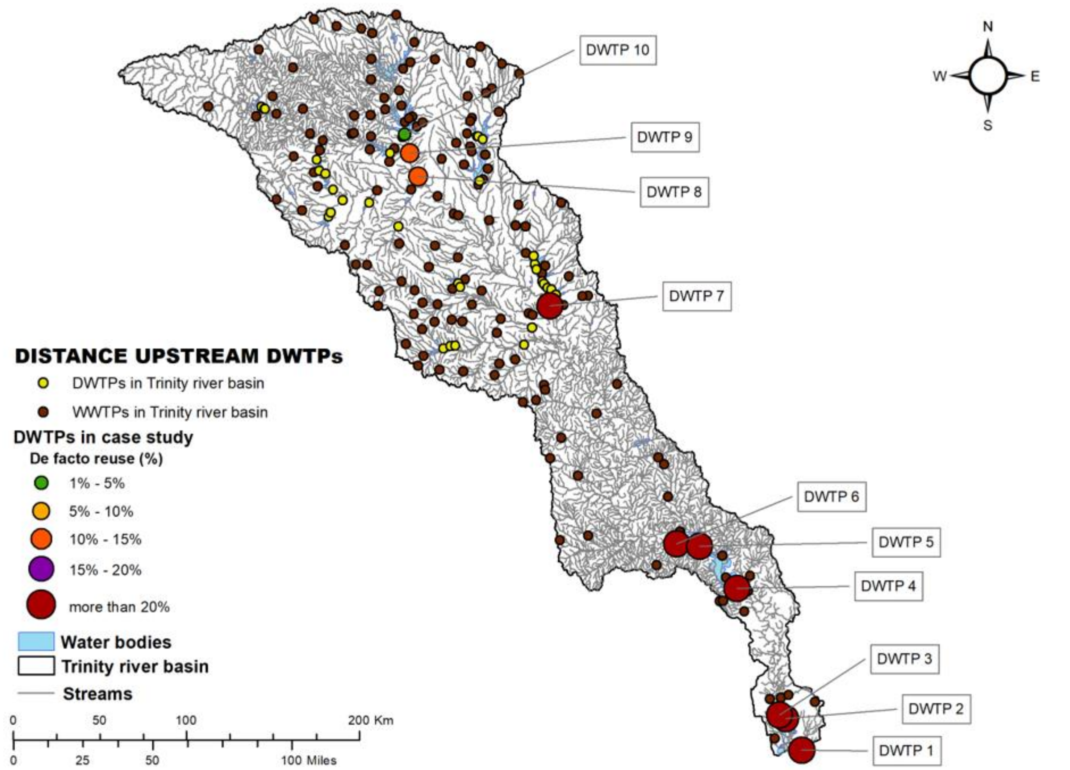


Figure 5.4 Locations of Ten DWTPs Used as a Case Study Impacted by 151 Upstream WWTPs in the Trinity River Basin

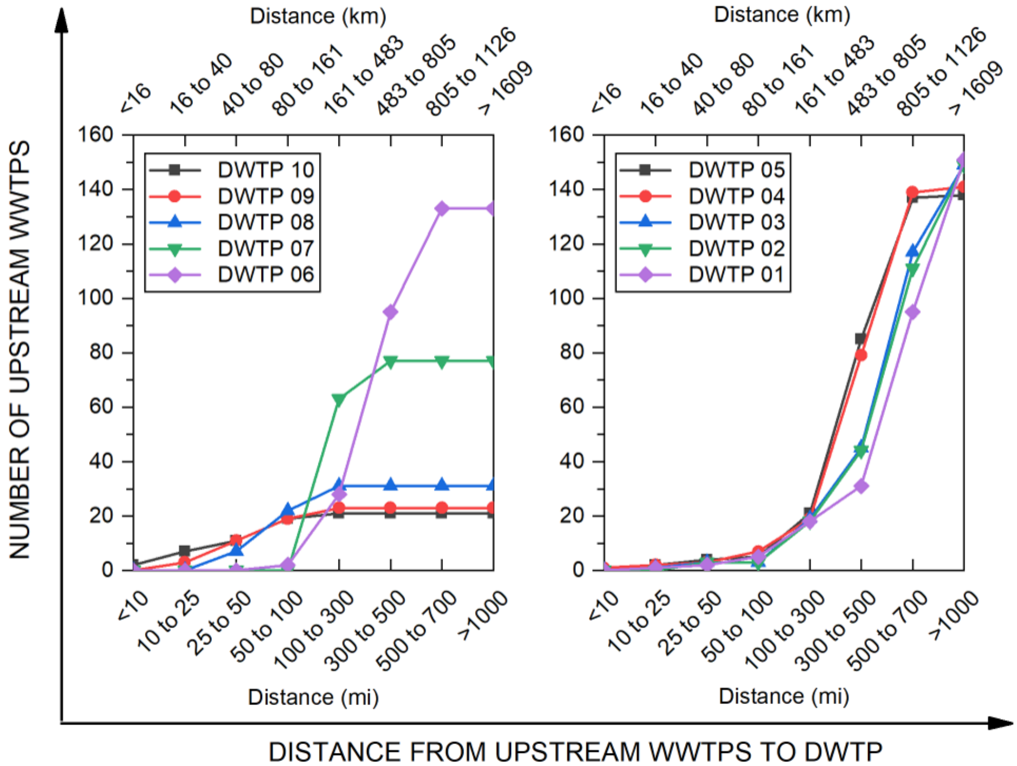


Figure 5.5. Proximity Analysis of Ten DWTPs Using Surface Water in the Trinity River Basin (in terms of Number of WWTPs Upstream)

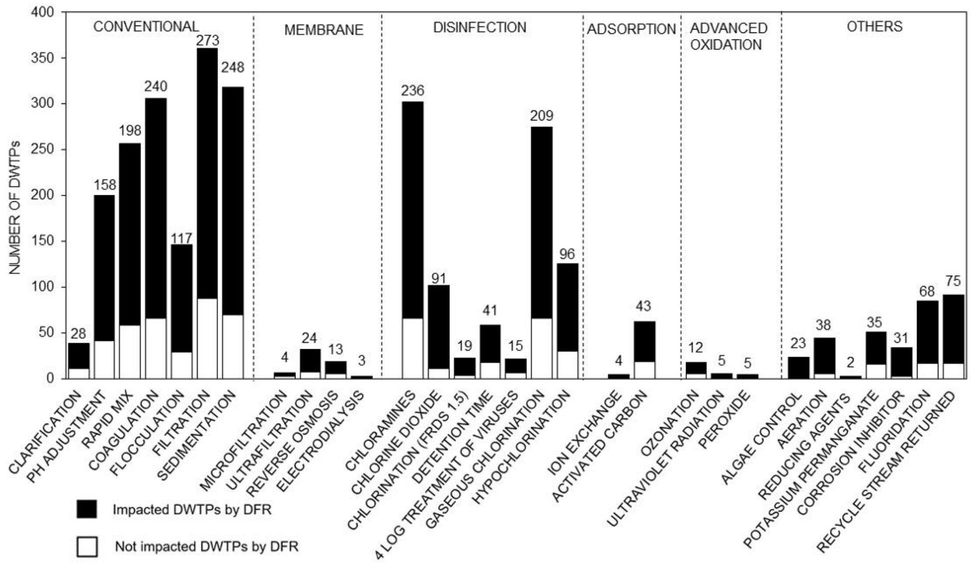


Figure 5.6 Unit Processes of DWTPs Using Surface Water in Texas (the Number Above Each Bar Represents the Number of DWTPs that are Impacted by DFR and Which Implement that Specific Type of Unit Process)

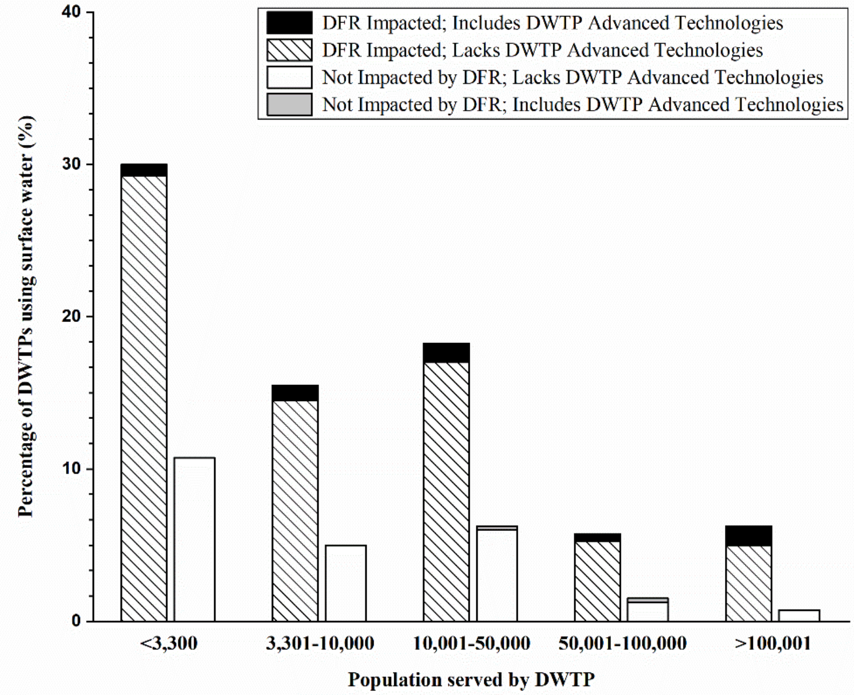


Figure 5.7 Percentage of Surface Water DWTPs in Texas Categorized by Population Served.

The percentage of non-impacted or impacted DWTPs using advanced technology was calculated as the number of DWTPs in each of four categories divided by the total number of DWTPs in Texas. Advanced technology is defined as using ozonation or with hydrogen peroxide granular activated carbon, or reverse osmosis.

Supporting Information

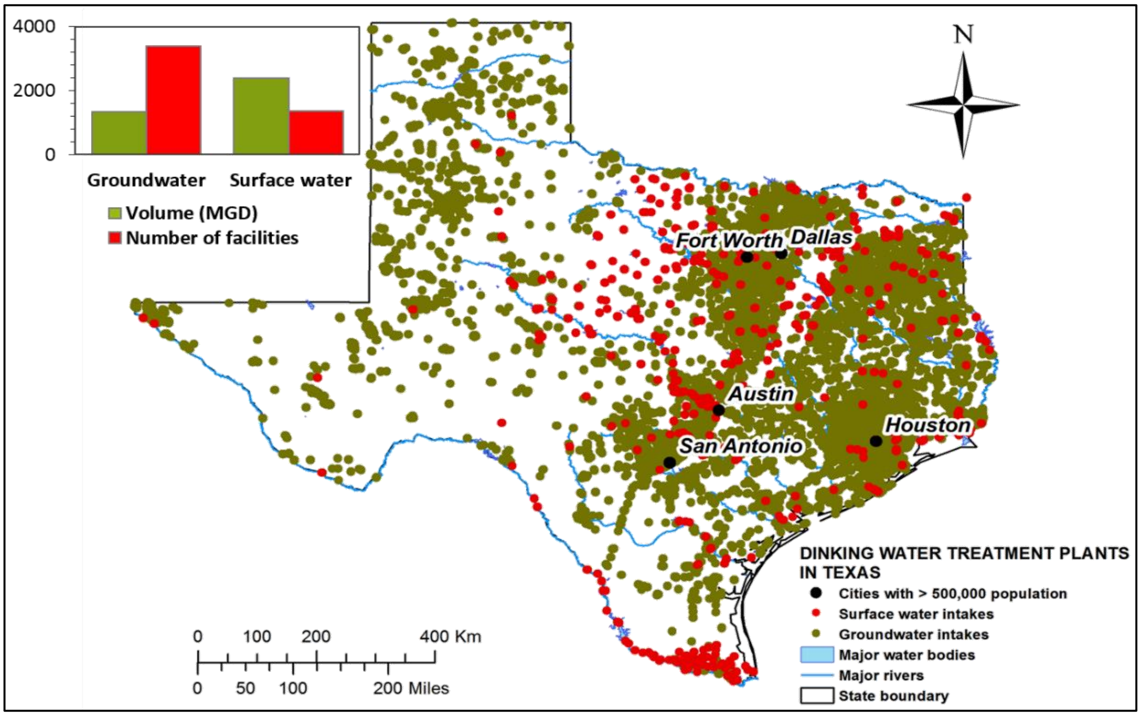


Figure SI. 1 Source Water for Drinking Water Facilities in Texas

A base layer for urban areas and main rivers in Texas obtained from the national atlas and the United States Geologic Surveys (USGS) was created using ArcGIS. The largest city in Texas is Houston with approximately 2.2 million residents, followed by San Antonio and Dallas each with an estimated population of ~1.2 million. Austin (the state capital), Fort Worth, El Paso, Arlington, Corpus Christi, Plano, and Laredo round out the top 10 urban population centers. Most urban areas are located in the eastern part of Texas and are typically near major rivers. As seen in Figure SI.1, there were 4,644 active DWTPs in Texas as of 2007. Approximately 73% (3,390 facilities) were groundwater intakes and 37% (1,254 facilities) were surface water intakes. According to the Historical Water Use Survey, 2010 - Texas Water Development Board, the reported annual municipal water use for Texas in 2010 was 4,204-acre feet (3,753 MGD).

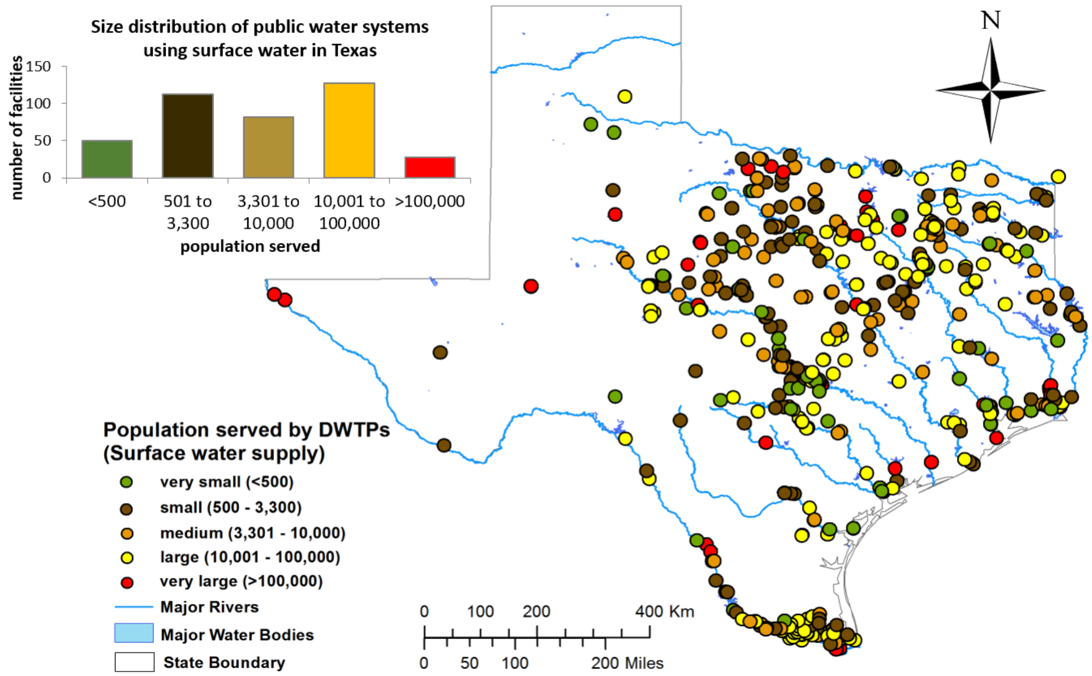


Figure SI. 2 Sizes and Locations of DWTPs Withdrawing Surface Water in Texas

Figure SI.2 (created in ArcGIS™ 10.4) shows the location of surface water intakes in Texas with the population served for all DWTPs. The map inset displays a histogram of DWTP sizes according to EPA's categories for the population served. Of 400 drinking water utilities, the number of very large DWTPs (serving more than 100,000 people) accounted for 7% (28/400). For instance, some very large DWTPs use surface water from Trinity River and Lake Houston to supply water for populated cities as Houston and Dallas. About two-thirds of DWTPs using surface water in Texas serve fewer than 10,000 people.

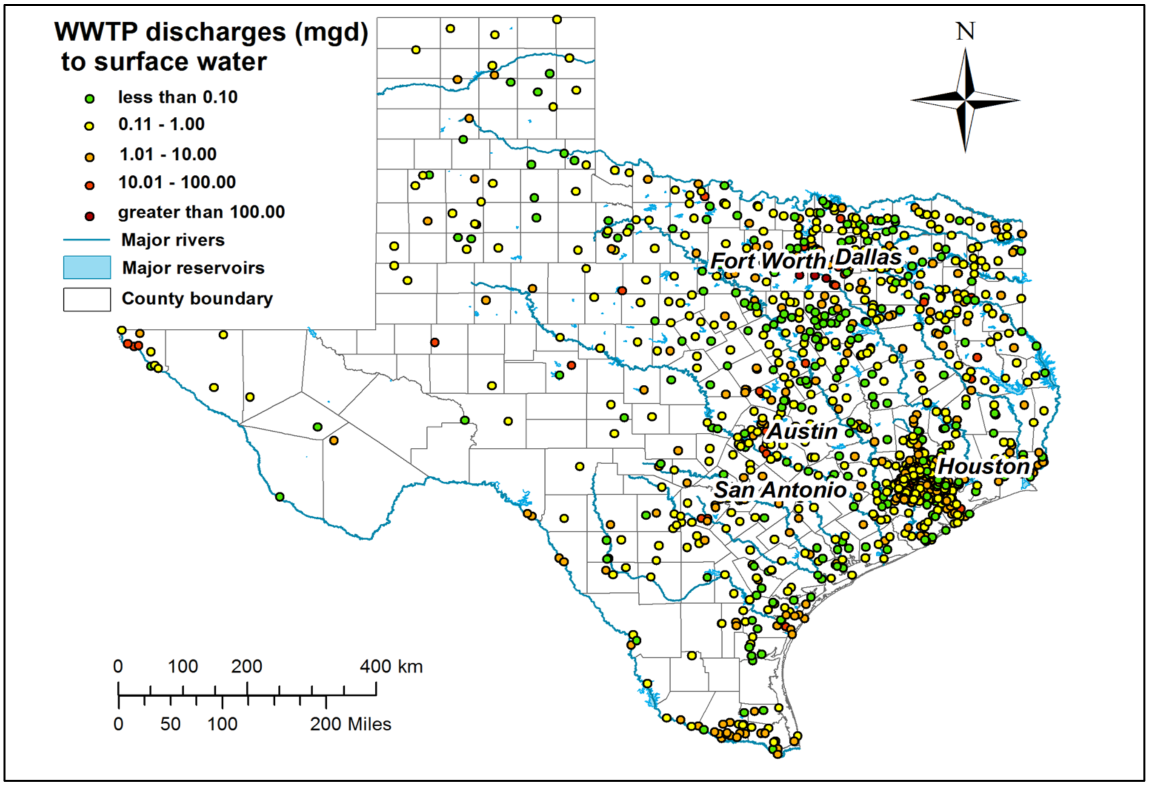


Figure SI. 3 Wastewater Discharge Outfalls to Surface Water in Texas

1,459 wastewater facilities with NPDES permits to discharge into surface water, groundwater, evaporation, spray for irrigation, and other locations in Texas. These facilities account for about 3,347 MGD of treated wastewater. About 85% (1,206/1,459 facilities) of WWTPs released 3,213 MGD of treated wastewater into surface water outfalls (96% of total treated wastewater volume) in Texas during 2008. Only about 15% of WWTPs disposed of wastewater effluent to other locations such as groundwater or ocean outfalls.

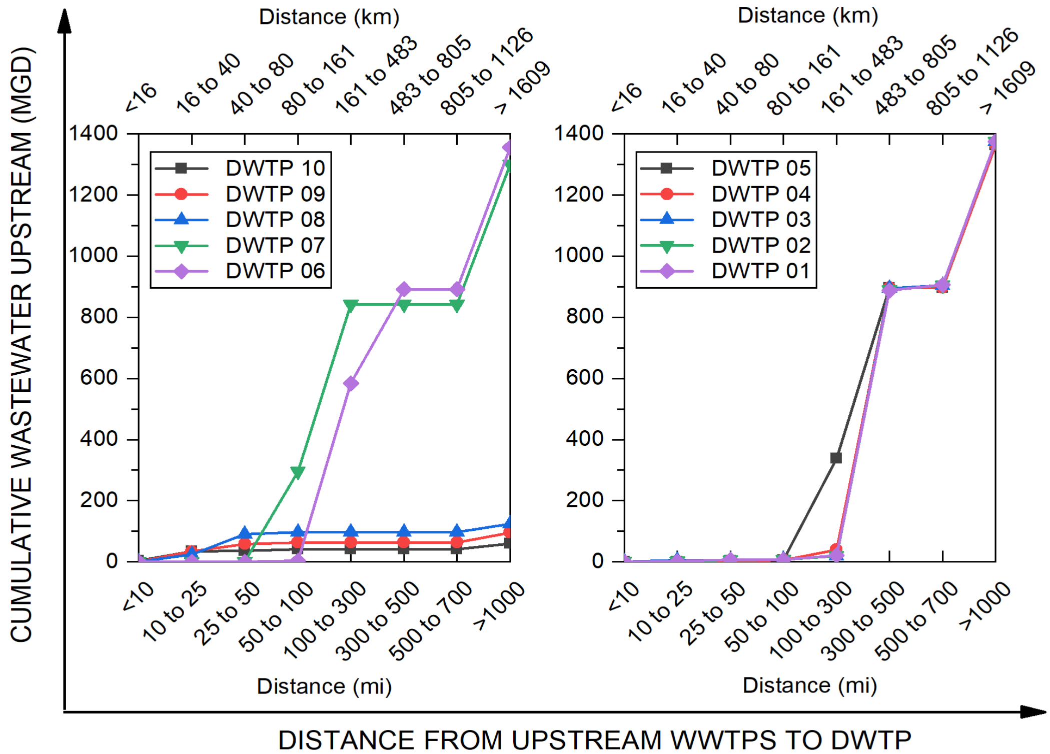


Figure SI. 4 Proximity Analysis on the Cumulative Wastewater Upstream to DWTPs in the Trinity River basin in Texas. Red, Blue and Green Lines for DWTP 04, DWTP 03, DWTP 02, respectively, are under the Purple Line for DWTP 01 due to a Similar Number

Table SI. 1 Cumulative Wastewater Versus Distance Between Upstream WWTPs and Downstream DWTPs

Distance (km)	Distance (mile)	DWTP 01	DWTP 02	DWTP 03	DWTP 04	DWTP 05
<16	<10	0.00	0.00	0.00	0.20	0.00
16 to 40	10 to 25	1.58	2.00	4.80	0.60	0.81
40 to 80	25 to 50	6.38	4.80	4.80	1.41	4.52
80 to 161	50 to 100	6.38	7.97	7.97	6.13	5.68
161 to 483	100 to 300	20.97	19.39	19.67	40.10	337.42
483 to 805	300 to 500	887.84	890.23	894.77	896.48	896.58
805 to 1126	500 to 700	906.53	904.95	904.95	897.18	896.58
> 1609	> 1000	1375.75	1375.52	1375.29	1363.74	1362.81

Distance (km)	Distance (mile)	DWTP 06	DWTP 07	DWTP 08	DWTP 09	DWTP 10
<16	<10	0.00	0.00	0.00	0.00	4.17
16 to 40	10 to 25	0.00	0.00	24.15	33.67	33.52
40 to 80	25 to 50	0.00	0.00	90.17	58.18	35.71
80 to 161	50 to 100	3.26	294.89	97.09	63.64	40.64
161 to 483	100 to 300	583.95	842.23	97.44	63.64	40.64
483 to 805	300 to 500	892.07	842.23	97.44	63.64	40.64
805 to 1126	500 to 700	892.07	842.23	97.44	63.64	40.64
> 1609	> 1000	1355.82	1300.61	123.67	95.58	60.00

Table SI. 2 Proximity Analysis of the number of WWTPs Upstream to DWTP in Trinity River Basin, Texas

Distance (km)	Distance (miles)	DWTP 01	DWTP 02	DWTP 03	DWTP 04	DWTP 05
<16	<10	0.00	0.00	0.00	1.00	0.00
16 to 40	10 to 25	2.00	1.00	3.00	3.00	2.00
40 to 80	25 to 50	5.00	3.00	3.00	5.00	5.00
80 to 161	50 to 100	5.00	9.00	9.00	10.00	9.00
161 to 483	100 to 300	29.00	27.00	29.00	63.00	71.00
483 to 805	300 to 500	123.00	130.00	133.00	140.00	138.00
805 to 1126	500 to 700	151.00	149.00	149.00	141.00	138.00
> 1609	>1000	151.00	149.00	149.00	141.00	138.00

Distance (km)	Distance (miles)	DWTP 06	DWTP 07	DWTP 08	DWTP 09	DWTP 10
<16	<10	0.00	0.00	0.00	0.00	4.00
16 to 40	10 to 25	0.00	0.00	1.00	8.00	9.00
40 to 80	25 to 50	0.00	0.00	17.00	14.00	15.00
80 to 161	50 to 100	6.00	9.00	29.00	23.00	21.00
161 to 483	100 to 300	74.00	77.00	31.00	23.00	21.00
483 to 805	300 to 500	133.00	77.00	31.00	23.00	21.00
805 to 1126	500 to 700	133.00	77.00	31.00	23.00	21.00
> 1609	>1000	133.00	77.00	31.00	23.00	21.00

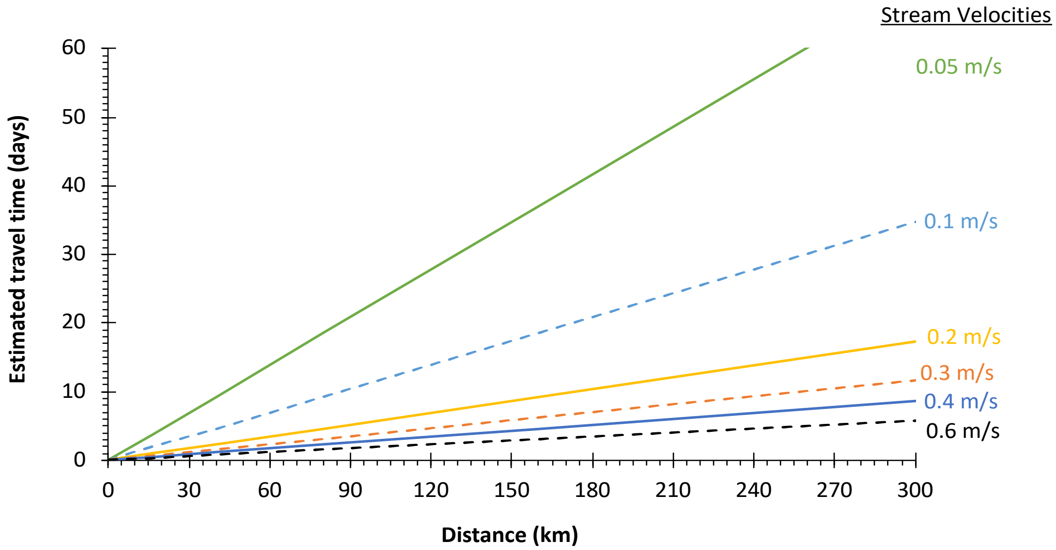


Figure SI. 5 Relationship between Proximal Distances WWTP discharges and DWTP Intakes and Estimated Travel Time Between the Two Points, for Streamflow Velocities Ranging from 0.05 to 0.6 m/sec

Table SI. 3 CEC Degradation Rates in Water from Fugacity Model

Chemicals	CAS	Half-life (hrs)	Half-life (days)	First-order rate constant, k (day-1)
TCPP	14609-54-2	1440	60	0.01155
TCEP	115-96-8	1440	60	0.01155
Diclofenac	15307-86-5	900	37.5	0.01848
Ibuprofen	15687-27-1	360	15	0.0462
Meprobamate	57-53-4	900	37.5	0.01848
Gemfibrozil	25812-30-0	900	37.5	0.01848
Sulfamethoxazole SMX	723-46-6	900	37.5	0.01848

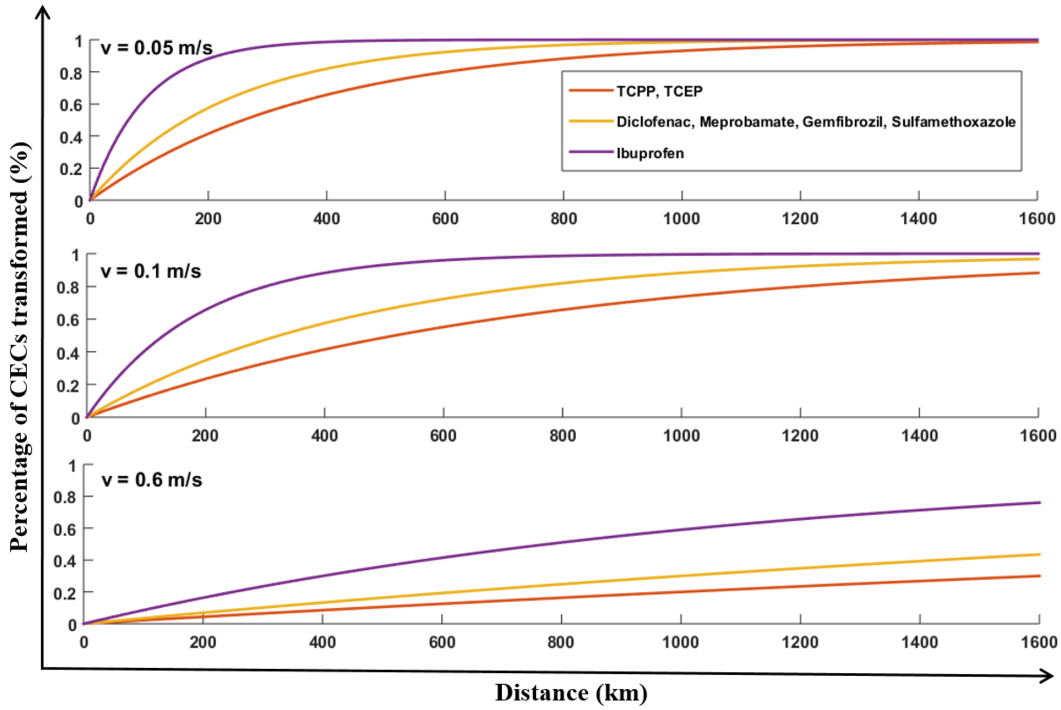


Figure SI. 6 The Percentage Transformation of 7 WWTP Surrogate CECs in Streams with different Streamflow Velocities (0.05 to 0.6 m/sec) Over Various Proximal Distances between WWTP Discharges and DWTP Intakes. Equation 1 was used to Estimate Transformations in Conjunction with Pseudo-first order rate Constants Summarized in Table SI.4

Table SI. 4 Unit Processes Installed for all DWTPs using Surface Water in Texas

Unit processes of drinking water treatment		Impacted by DFR	Not impacted by DFR
		Total	Total
CONVENTIONAL	CLARIFICATION	28	11
CONVENTIONAL	PH ADJUSTMENT	158	42
CONVENTIONAL	RAPID MIX	198	59
CONVENTIONAL	COAGULATION	240	66
CONVENTIONAL	FLOCCULATION	117	29
CONVENTIONAL	FILTRATION	273	88
CONVENTIONAL	SEDIMENTATION	248	70
MEMBRANE	MICROFILTRATION	4	3
MEMBRANE	ULTRAFILTRATION	24	8
MEMBRANE	REVERSE OSMOSIS	13	6
MEMBRANE	ELECTRODIALYSIS	3	0
DISINFECTION	CHLORAMINES	236	66
DISINFECTION	CHLORINE DIOXIDE	91	11
DISINFECTION	CHLORINATION (FRDS 1.5)	19	4
DISINFECTION	DETENTION TIME	41	18
DISINFECTION	4 LOG TREATMENT OF VIRUSES	15	7
DISINFECTION	GASEOUS CHLORINATION	209	66
DISINFECTION	HYPOCHLORINATION	96	30
ADSORPTION	ION EXCHANGE	4	1
ADSORPTION	ACTIVATED CARBON	43	19
ADVANCED OXIDATION	OZONATION	12	6
ADVANCED OXIDATION	ULTRAVIOLET RADIATION	5	1
ADVANCED OXIDATION	PEROXIDE	5	0
OTHERS	ALGAE CONTROL	23	1
OTHERS	AERATION	38	6
OTHERS	REDUCING AGENTS	2	1
OTHERS	POTASSIUM PERMANGANATE	35	16
OTHERS	CORROSION INHIBITOR	31	3
OTHERS	FLUORIDATION	68	17
OTHERS	RECYCLE STREAM RETURNED	75	17

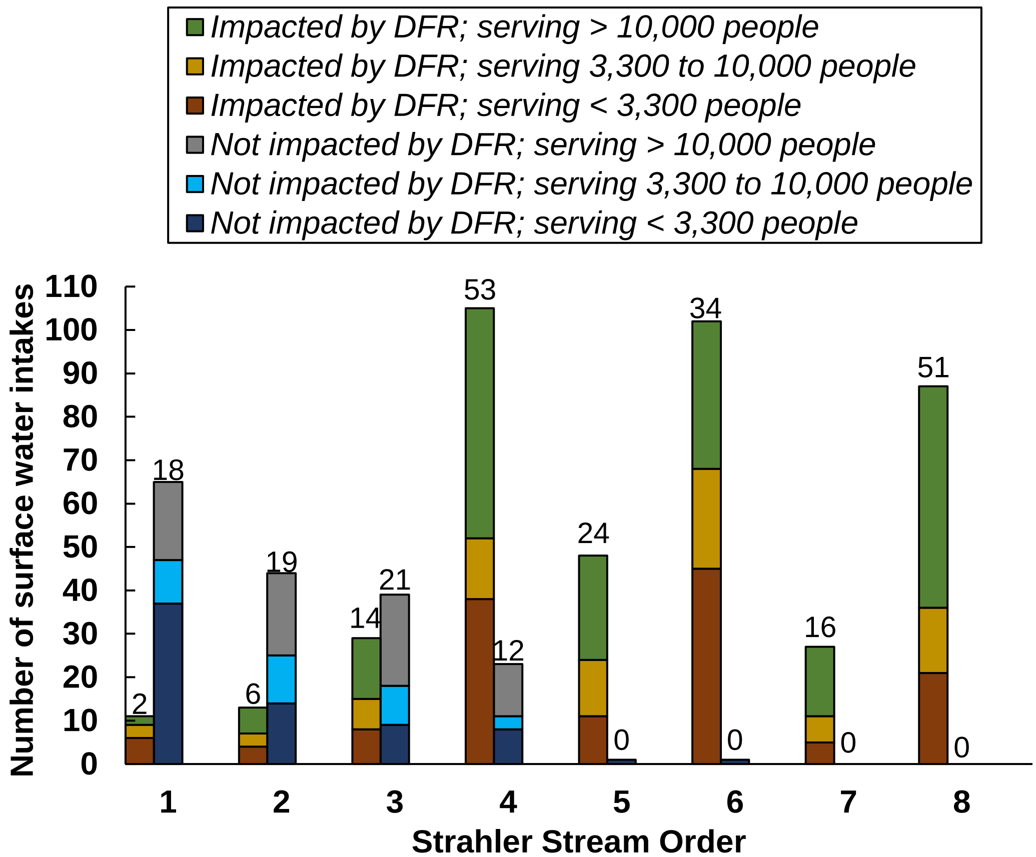


Figure SI. 7 Distribution of Surface Water intakes in Texas Categorized by Population served and Strahler Stream Order (the number for each Bar Indicates for Surface Water Intakes belong to DWTPs serving 10,000 or more people)

Table SI. 5 Number of Surface Water Intakes vs Stream Order (categorized by population served)

	Impacted by DFR				Not impacted by DFR			
	Population served by DWTPs				Population served by DWTPs			
Order	<3,300 people	3,300 to 10,000 people	>10,000 people	Tot al	<3,300 people	3,300 to 10,000 people	>10,000 people	Total
1	6	3	2	11	37	10	18	65
2	4	3	6	13	14	11	19	44
3	8	7	14	29	9	9	21	39
4	38	14	53	105	8	3	12	23
5	11	13	24	48	1	0	0	1
6	45	23	34	102	1	0	0	1
7	5	6	16	27	0	0	0	0
8	21	15	51	87	0	0	0	0
Total	138	84	200	422	70	33	70	173

Table SI. 6 Number of violations at DWTPs using surface water in Texas (total 316 facilities; 246 systems with violations (3 years), 194 of these 246 systems are reported with violations)

	Number of violations*		DWTPs with violations	
Population served by DWTPs	Number	Percentage (%)	Facility	Percentage
<3.3k	282	47.72	79	40.72
3.3k to 10k	181	30.63	53	27.32
>10k	128	21.66	62	31.96
Total	591	100	194	100

Number of violations*: a DWTP can have more than one non-compliance violation.

Table SI. 7 Number of health-based violations at DWTPs using surface water in Texas

	Impacted DWTPs		Non-Impacted DWTPs	
Population served	DFR impacted with health-based violations	DFR impacted without health-based violations	Non-Impacted DWTPs with health-based violations	Non-Impacted DWTPs without health-based violations
< 3,300	32	89	10	32
3,301-10,000	29	34	8	11
10,001 - 50,000	22	48	6	22
50,001 - 100,000	4	20	1	4
>100,001	4	21	0	3
Total	91	212	25	72
	303		97	

Figure SI. 8 Top 10 MCL violations at DWTPs using surface water in Texas (total of 194 DWTPs with violations)

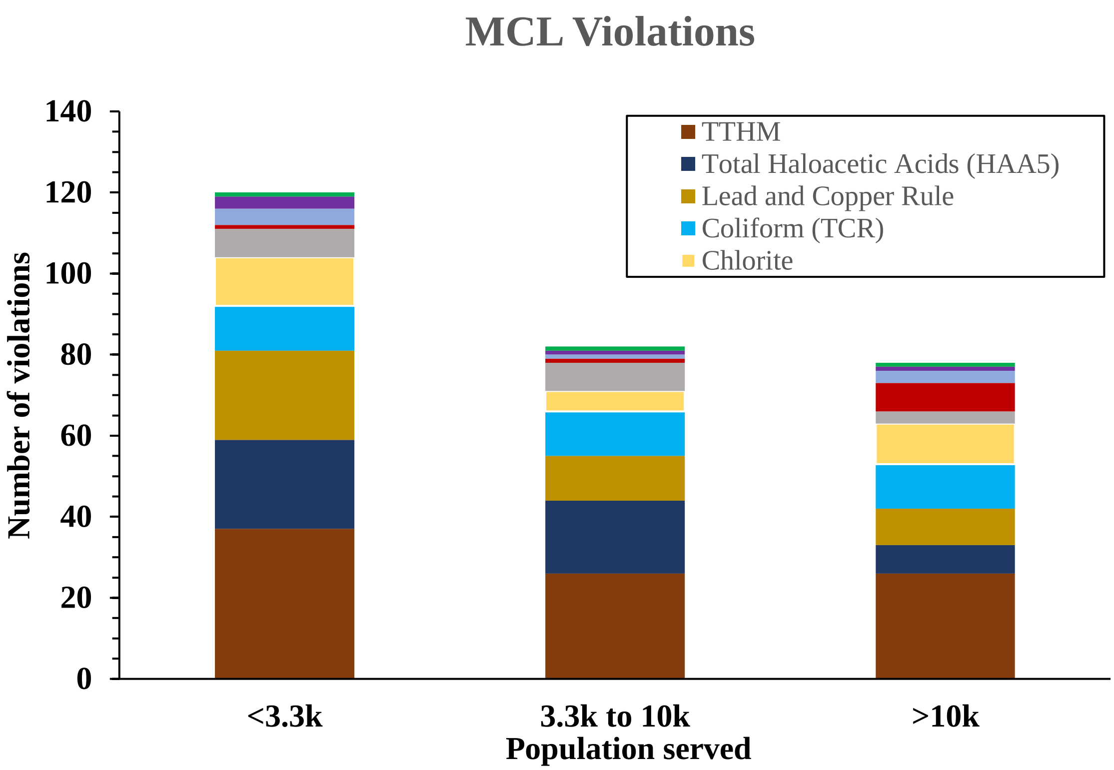
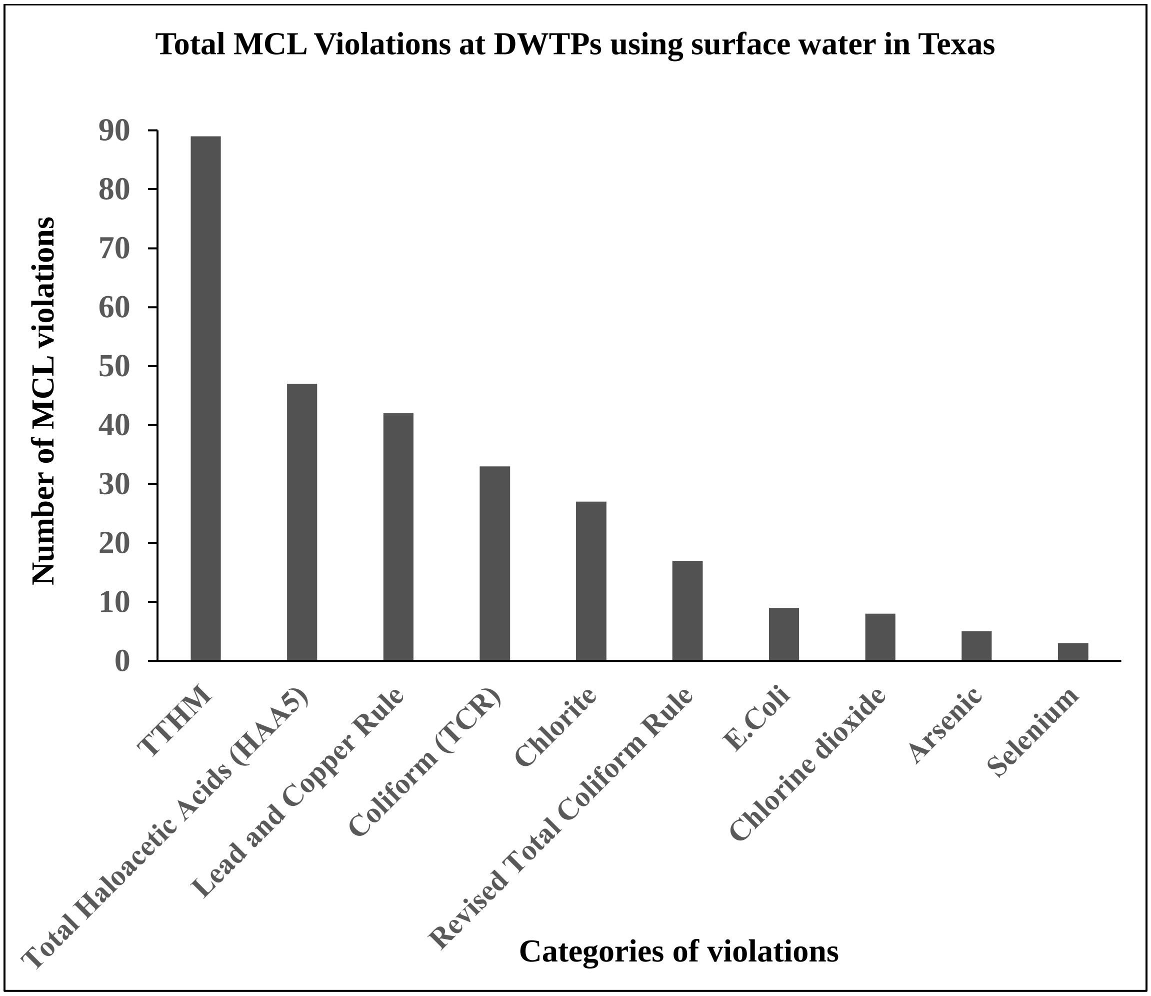


Figure SI. 9 Top 10 MCL violations at DWTPs using surface water in Texas categorized by population served (a DWTP can have more than one non-compliance violation)



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CHAPTER 6

UNPLANNED WATER REUSE IMPACTS ON SURFACE WATER SOURCES FOR SMALLER-VERSUS LARGER WATER SYSTEMS ACROSS THE UNITED STATES

This chapter has not been published yet. The manuscript is prepared to submit to the Environmental Health Journal. Contributing authors are Thuy Nguyen, Julianna Acero, and Paul Westerhoff*.

My author's contribution is to develop the new feature of the model, derive the model results, analyze the data, and lead the writing of the manuscript.

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Target Journal: *Environmental Health Journal*

Keywords: de facto reuse, small public water system, treated wastewater, serving fewer than 10,000 people, drinking water quality

Abstract

Small water systems generally have less technical, management, and financial capacity than larger water systems to comply with the Safe Drinking Water Act standards. Potential risks of small water systems to the contaminant of emerging concerns (CECs) originating from treated wastewater in finished drinking water (i.e., de facto reuse) may be more significant than large water systems. Yet very little data is available to estimate the occurrence of de facto reuse associated with smaller water systems, this study conducted to quantify and compare the impact of de facto reuse in surface water sources at smaller water systems (serving $\leq$ 10,000 people) versus larger water systems (serving>10,000 people) across the United States. The hypothesis tested whether smaller DWTPs in the United States is disproportionally dependent upon treated wastewater and lack advanced technology capable of removing CECs in surface water sources. To achieve the goal, the DRINCS model was expanded to include smaller DWTPs serving 10,000 or fewer people ($N = 6,045$ surface water intakes at 3,984 DWTPs). Nationally, > 40% of SW intakes at all DWTPs across the U.S. were impacted by DFR under average flow ($N = 2,917$ of 6,826). Smaller DWTPs had a higher frequency of DFR than larger DWTPs with 1,504 and 1,413, respectively. However, the difference in the level of DFR at smaller versus larger DWTPs was statistically unclear (t-test, $p = 0.274$). Smaller communities relied on SW from a low order stream impacted by DFR together with lacking a doubled number of advanced DWTP unit processes than larger systems could have high risks to CECs. Levels of DFR for larger DWTPs were statistically higher on mid-size stream orders than those for smaller systems. As they serve large communities, this could pose risks to a population as 40 times as those served by smaller systems. The total exposed population to the risks of CECs to source water was estimated at 73 million (at DFR>1%) and 12.3 million people (at DFR>10%). Future studies can use DFR results to conduct epidemiological and risk assessment studies for communities impacted by CECs and identify communities that would benefit from advanced treatment processes that remove CECs.

Introduction

Unplanned (de facto) potable water reuse is defined by the National Research Council as "a drinking water supply that contains a significant fraction of wastewater effluent, typically from upstream wastewater discharges, although the water supply has not been permitted as a water reuse project" (Reuse, 2012). In the United States, over 16,000 POTWs were discharging more than 70% of treated wastewater (or 21.3 billion gallons per day out of 27.1 billion gallons per day) into surface water (USEPA, 2012). More than half of this number of POTWs employed conventional (or secondary) wastewater treatment which is insufficient to remove numerous contaminants in raw wastewater completely (Metcalf et al., 2007). Treated wastewater effluents are major point sources of contaminants of emerging concerns (CECs) (e.g., pathogens, organic precursors, disinfection by-products, pharmaceuticals) into surface water sources (Reuse, 2012). For example, pathogen *Cryptosporidium* has been reported in secondary effluents in the United States with a range of 0.1 and 40.8 oocysts/liter (McCuin & Clancy, 2006). Protozoa agents (*Cryptosporidium, Giardia, Microsporidia*) can cause acute gastrointestinal illness and potentially disease outbreaks (Metcalf et al., 2007). Disinfection by-products (DBPs) can be formed in the excess presence of organic matter, nitrogen, iodine, and bromide in municipal wastewater effluents during the disinfection process (Stuart W. Krasner, Westerhoff, Chen, Rittmann, & Amy, 2009). Trussell (1978) reported the formation of halogenated DBPs (e.g., trihalomethanes THMs and haloacetic acids HAAs) in chlorination-treated waters. Ozonation can form nitrogenous, iodinated and, brominated DBPs (Huang, Fang, & Wang, 2005). A non-halogenated DBPs, N-nitrosamine was found during the chloramination process (Hanigan et al., 2015; Stuart W Krasner, 2009; Mitch et al., 2003), and reported in drinking water in compliance with the Stage 2 Disinfectants and Disinfection By-Products Rule (Seidel, McGuire, Summers, & Via, 2005). Haloacetonitriles (HANs) and haloacetaldehydes (HALs) are identified as important unregulated-DBPs groups while iodinated and brominated DBPs are among the most genotoxic of those currently found in water (Richardson et al., 2008). Surface water sources are vulnerable to a higher level of CECs which can increase the potential risks to human health in drinking water.

More than 70% of the U.S. population (or over 220 million people) are supplied by public water systems using surface water sources (e.g., lakes, rivers, reservoirs) (Dieter et al., 2018). Advanced unit processes at a surface water treatment plant employ activated carbon, advanced oxidation process, ozonation, ultraviolet irradiation, membrane (reverse osmosis or nanofiltration) can be capable of removing CECs in source waters (Reuse, 2012). Ultraviolet irradiation has been found effective for removal/inactivating G.*lamblia* and G.muris (Howe, Hand, Crittenden, Trussell, & Tchobanoglous, 2012); and *Cryptosporidium* oocysts (Craik, Weldon, Finch, Bolton, & Belosevic, 2001). Two main size categories in this study are smaller public water systems (serving ≤ 10,000 people) and larger public water systems (serving > 10,000 people) based on the number of the population served (USEPA, 2019b).

Compared to larger water systems (DWTPs), smaller water systems generally have less technical, management, and financial capacity to comply with Safe Drinking Water Act standards (USEPA, 1996). A summarized statistics of violations for all sized DWTPs in the U.S reported that health-violations were likely to occur at smaller DWTPs (serving between 501 and 10,000 people) than at larger water systems (Rubin, 2013). A De facto Reuse Incidence in our Nation's Consumable Supply (DRINCS) model, previously developed by Rice and Westerhoff (2015) and confirmed by T. Nguyen et al. (2018), estimated levels of de facto reuse for 2,056 surface water sources at 1,210 of the largest DWTPs (serving >10,000 people) in the United States. The modeled results predicted a high frequency of DFR at more than 50% of drinking water intakes for these larger water systems but with a relatively low magnitude of less than 1% DFR under mean annual streamflow condition (Rice, Via, & Westerhoff, 2015; Rice & Westerhoff, 2015). The DRINCS model has been expanded to evaluate additional smaller DWTPs (325 surface water intakes at 245 smaller DWTPs) into an existing of 270 surface water intakes at 155 larger DWTPs in the previous model (T. T. Nguyen & Westerhoff, 2019). The modeled results for Texas predicted that two-thirds of all surface water intakes were impacted by at least one WWTP upstream. The level of DFR at SW intakes in Texas ranged between 1 to 20% under average streamflow and exceeded 90% during mild droughts. Smaller DWTPs in Texas had a higher frequency of DFR than larger systems while fewer than 10% of these DWTPs employed advanced technologies capable of removing CECs (T. T. Nguyen & Westerhoff, 2019). Nationally, compared to larger DWTPs, there are nearly three times as many surface water intakes at smaller DWTPs serving ≤ 10,000 people as those at larger systems (N = 6,045 surface water intakes at 3,984 smaller DWTPs) across the United States (USEPA, 2019c). Globally, several studies quantified a national assessment of de facto reuse in surface water supplies (Drewes, Hübner, Zhiteneva, & Karakurt, 2017; MLIT, 2019). In Switzerland, a nationwide assessment estimated DFR under dry weather flow (Q347) conditions and many streams in the populated northern part of Switzerland had more than 20% DFR (Drewes et al., 2017). The river reaches in Germany were estimated to constitute more than 30-50% treated wastewater effluents under mean minimum discharge conditions between May and September of the year (Karakurt, Schmid, Hübner, & Drewes, 2019). A nationwide survey on Japanese rivers has reported the Freshness Level of the wastewater effluent receiving rivers with the range between 21% and 73% for two years of 2003 and 2004 (MLIT, 2019).

Yet very little data is available to estimate de facto reuse associated with these systems, the risks of smaller public water systems to CECs of wastewater origin in finished drinking water may be more significant than larger systems. This study expands the DRINCS model to include smaller DWTPs (serving 10,000 or fewer people) with fully ground-truthed locations of DWTPs and WWTPs and compare impacts of de facto reuse on surface water sources for smaller and larger water systems.

Methods

Wastewater treatment plants (WWTPs) were obtained from the EPA Clean Watersheds Needs Survey (CWNS 2012), released in May 2016, including WWTP facility information, design capacity, level of treatment, and coordinates of wastewater outfalls to surface water (N = 16,161) (USEPA, 2016). The expanded DRINCS utilizing DWTP data from the Safe Drinking Water Information System (SDWIS/Federal, the calendar year 2019) included 6,826 active surface water intakes at 3,947 DWTPs for all 48 contiguous states in the US, District Columbia, and the tribal regions (USEPA, 2019c). An active public water system operates regularly or seasonally within the year while inactive water systems go out of business or are abandoned or merged into another water system (USEPA, 2019b).

Additional information on stream segments receiving WWTP outfalls or supplying DWTP surface water intakes was collected from state databases or other electronic sources (e.g., the National Pollutant Discharges Systems (NPDES) permit program; the Consumer Confidence Report, and the Drinking Water Watch website). It is important to ground-truth the precise locations with a two-step approach using Google Earth visualization and checking the state data on stream segments. First, the locations of WWTP outfalls or DWTP surface water intakes were spatially joined to the closest stream segment in the ArcGIS™. Secondly, the attribute tables of the joined layer and the state database were compared in Microsoft Excel to identify the right stream segment with matched names of effluent receiving rivers or surface water supplies. Further, the SDWIS Federal database provides the DWTP unit processes at each location, the treatment objectives for all DWTPs in the DRINCS model. In this study, all the locations of WWTP outfalls (N = 16,161) and DWTP surface water intakes (N = 6,826) were fully ground-truthed.

The U.S Geological Survey National Hydrography Dataset Plus (NHDPlus V2) provides medium-resolution NHD Flowline features (1:100,000 scale) with more than three million stream segments and value-added attributes of mean annual streamflow, Strahler stream order, digital flow direction, and identification number of the river reaches. The ArcMap™ version 10.4, Python 2.7, and the Network Analysis extension were used in this study to perform the automation process, geoprocessing tools, and mapping visualization. The continental United States was divided into 12 Vector Processing Units (VPUs) (NHDPlus, 2012). For each VPU, there was a built geometric network with digitized flow directions and network analysis within the ArcGIS framework (T. Nguyen et al., 2018; T. T. Nguyen & Westerhoff, 2019). Python scripts were written to automate the calculation of the cumulative treated wastewater effluents upstream to a DWTP surface water intake at each VPU. De facto reuse is calculated from the cumulative of upstream WWTP design discharge flow ($Q_{WW, i}$) divided by the streamflow of the stream segment at surface water intakes (Q_{SW}) (Equation 1):

$$DFR = \frac{\sum Q_{WW,i}}{Q_{SW}} x100\% \qquad (1)$$

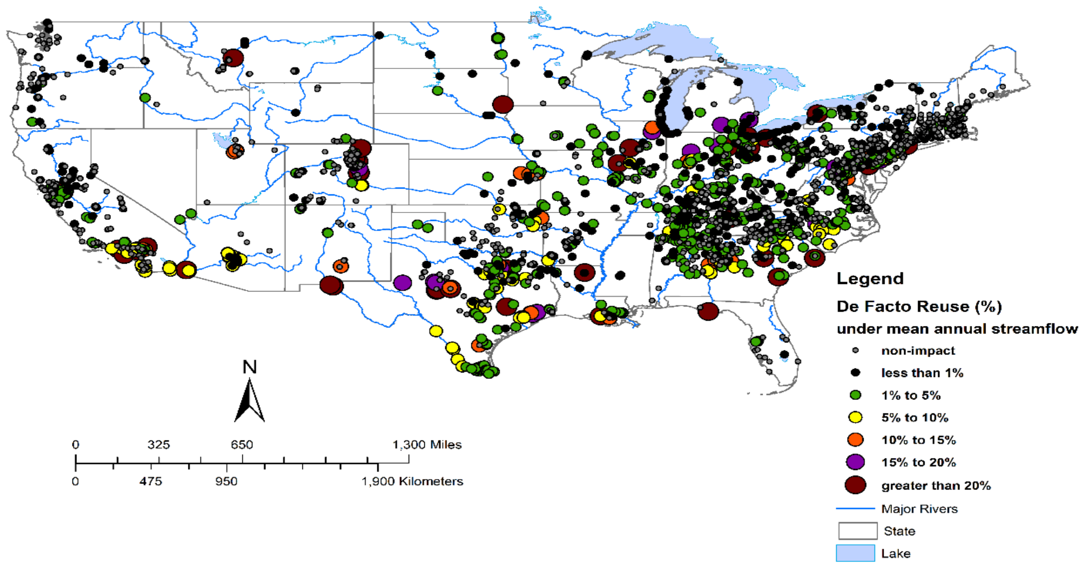
The Enhanced Unit Runoff Method (EROM) flow estimation was valid for the 1971 to 2000 time period. The locations of surface intakes were spatially joined with the closest USGS stream gages to derive historical stream flows. The 7Q10 (the 7 day lowest streamflow in a 10-year recurrence interval) was calculated by using “Basin 4” software developed by US EPA (USEPA, 2019a). The 7Q10 for ungagged streams were obtained from the StreamStats website (https://streamstats.usgs.gov/ss/). Statistical analyses were performed using Minitab™ Express Statistic Software. Welch’s two-sample t-test for unequal sample sizes of two groups tests the null hypothesis whether the difference between two mean values of DFR at smaller versus larger DWTPs **equals** by hypothesized difference. All three alternative hypotheses include the difference between means **does not equal to, greater than, or lower than** hypothesized difference.

Results and Discussions

Frequency of De Facto Reuse under Annual Mean Flow Condition

The DRINCS modeled de facto reuse at all surface water intakes at smaller DWTPs (N = 3,911 in Figure 6.1B) and larger DWTPs (N = 2,915 in Figure 6.11A) under annual mean streamflow condition. Nationally, more than 40% of surface water intakes at all sized DWTPs across the U.S. were impacted by de facto reuse under annual mean streamflow (N = 2,917 of 6,826). Smaller DWTPs had a higher number of surface water intakes impacted by DFR under annual mean streamflow compared with larger water systems with 22% (1,504 impacted SW intakes at 1,192 systems) and 20.7% (1,413 impacted intakes at 910 systems), respectively. Several rivers were predicted to receive treated wastewater up to 50% under annual mean flow conditions which included Trinity River, South Concho River, South Platte River, and Los Angeles River. Figure 6.1 shows a spatially explicit map of the de facto reuse level produced by using the DRINCS model with the updated and expanded data. The occurrence of de facto reuse was unevenly distributed across the U.S. while the high density of DFR was illustrated in areas of California River, Arkansas White Red, Mississippi River, and the Rio Grande basins.

A. Surface Water Intakes at Public Water Systems Serving more than 10,000 People



B. Surface Water Intakes at Public Water Systems Serving 10,000 or fewer People

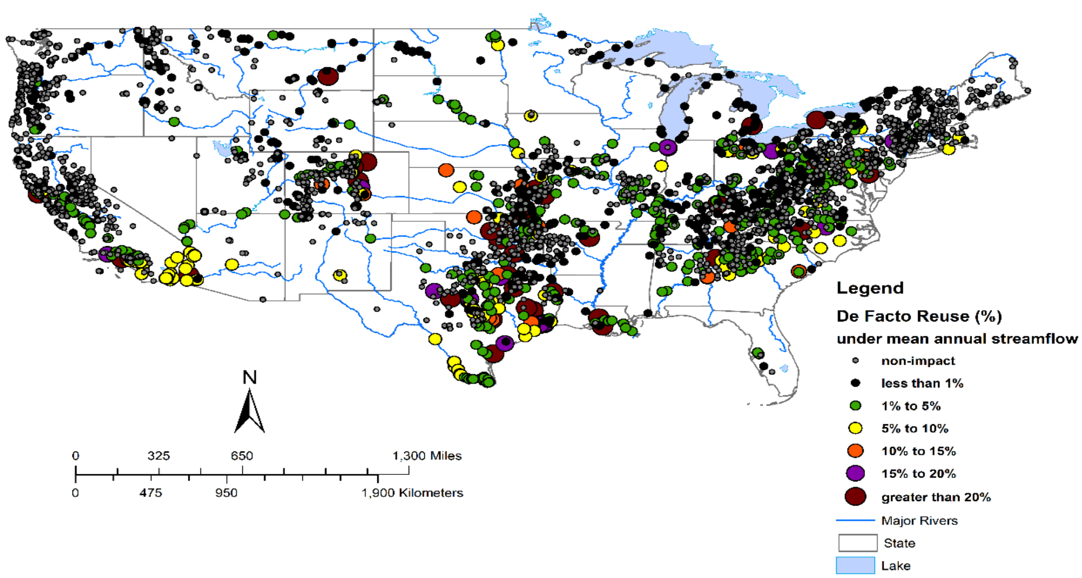


Figure 6.1 Geographical Distribution of De Facto Reuse at Surface Water Intakes Under Mean Annual Streamflow Condition. A) Large Surface Water Systems Serving >10,000 People; B) Small Surface Water Systems Serving ≤ 10,000 People

The Magnitude of De Facto Reuse under Annual Mean Flow Condition

The distribution of surface water intakes as a function of the DFR level for smaller versus larger DWTPs is displayed in Figure 6.2. As the magnitude of DFR increases, the number of surface water intakes impacted by DFR tends to decrease in two sized groups. It can be seen clearly in Figure 6.2 that smaller DWTPs had more surface water intakes not being impacted by treated WWTPs upstream. The lowest number of surface water intakes impacted by DFR was in the DFR range of 15% to 20%.

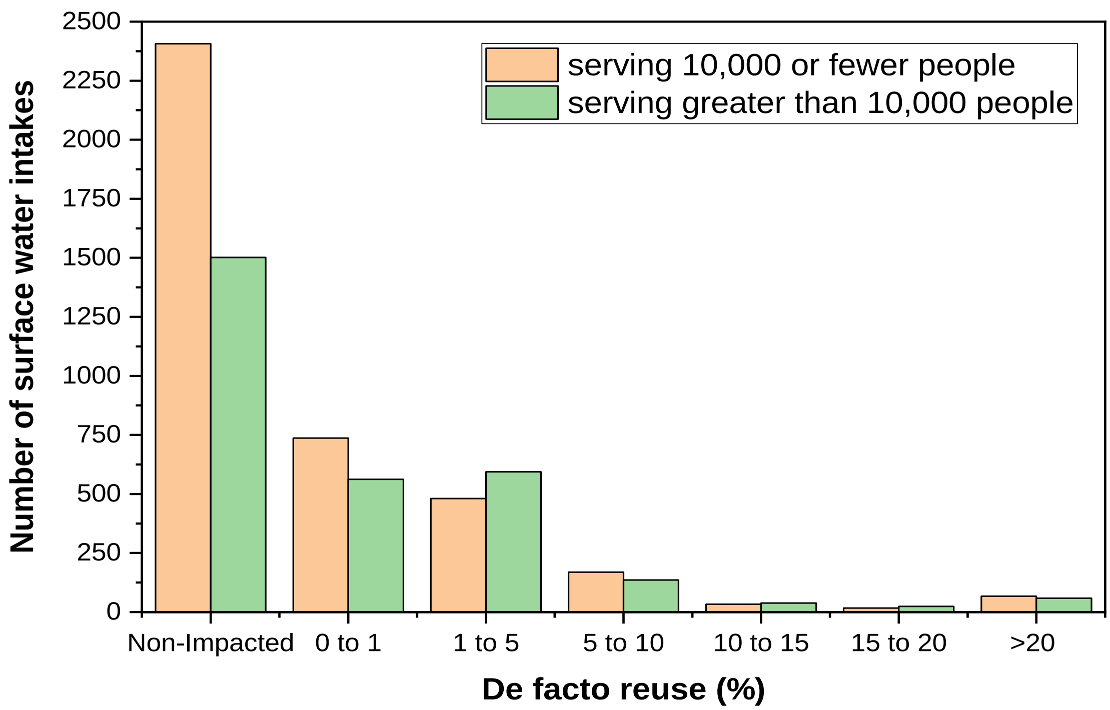


Figure 6.2 Distribution of Surface Water Intakes Categorized by Level of De Facto Reuse and Population Served

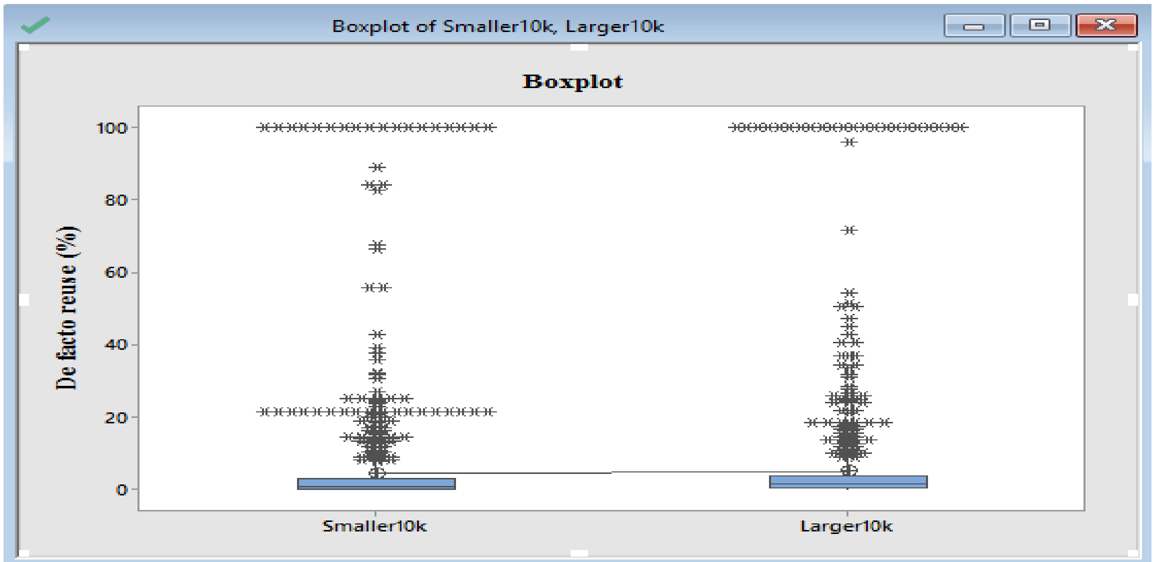


Figure 6.3 BoxPlot showing Variation in Levels of De Facto Reuse at Surface Water Intakes for Smaller Versus Larger DWTPs

The two-sample t-test was performed to determine if there is a significant difference between two means of the DFR level in smaller DWTPs and larger DWTPs. A null hypothesis for no difference. The alternative hypothesis is a significant difference between DFR means for smaller versus larger DWTPs (the significant level alpha is 0.05). The difference between the means of DFR in surface water at smaller vs larger DWTPs was statistically unclear (t-test, $t(2917) = -0.536$, $p = 0.274$).

The Magnitude of De Facto Reuse under 7Q10 Flow Condition

Temporal variations in dilution flows will affect surface water quality. Specific flow criteria (e.g., annual mean flow, 7Q10 (average low-flow over 7 consecutive days with a 10-year return interval) were used to evaluate the extent and significance of de facto reuse variation in surface water. 7Q10 is a hydrologically based designed flow developed by the U.S. Geological Survey and used in many states as a low flow condition for setting permit discharge limit (http://www.epa.gov/ceam). In the United States, 7Q10 is widely used as a low flow index and also indicates an extreme low flow condition exceeding the 90^{th} percentile flow (or P5 on the flow curve). The normal flow condition or the 50^{th} percentile is the flow that is equal to or exceeds 50% of the recorded flow values. Using an extensive database from the USGS Stream gauge stations, this study investigates the impact of varied flow conditions in the stream network on the variation on DFR. But, due to limited gauges stations to monitor million river segments in the U.S., only 274 stream gauges were available to obtain adequate long-term (>30 years) historical streamflow data.

Figure 6.4. illustrates the variation in DFR under low flow (7Q10) condition as a function of Strahler stream orders for smaller versus larger DWTPs. Mid-sized Strahler stream orders had a wide range of DFR variation (up to 100% of treated wastewater during low flow) while higher stream orders (>8) had a narrow variation. Except for small streams (Order 1 and 2) with only one available stream gauge, there are a significant number of stream gauges for larger streams.

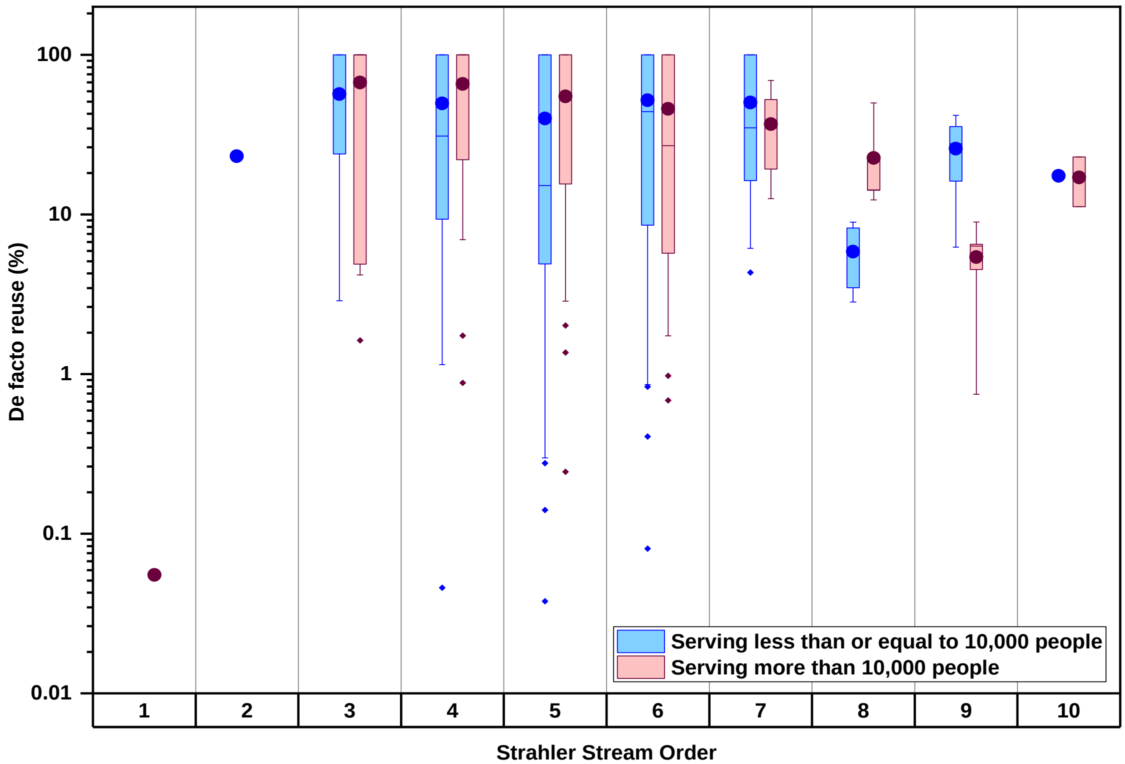


Figure 6.4 Boxplot of the Variation of DFR in Surface Water under Low Flow (7Q10) condition as a Function of Strahler Stream Order

The variation of treated wastewater in the receiving rivers (DFR) was examined and compared for under 7Q10 low flow and 50^{th} percentile average flow conditions. From the calculation, 52 of 274 (or 19%) sites had >10% DFR in the source water under the 50th percentile flow while 185 of 274 (or 68%) at the 7Q10 flow condition. A decrease in streamflow may lower the in-stream dilution and increase DFR in the receiving stream as the level of DFR was significantly higher under 7Q10 low flow than the 50^{th} percentile average flow.

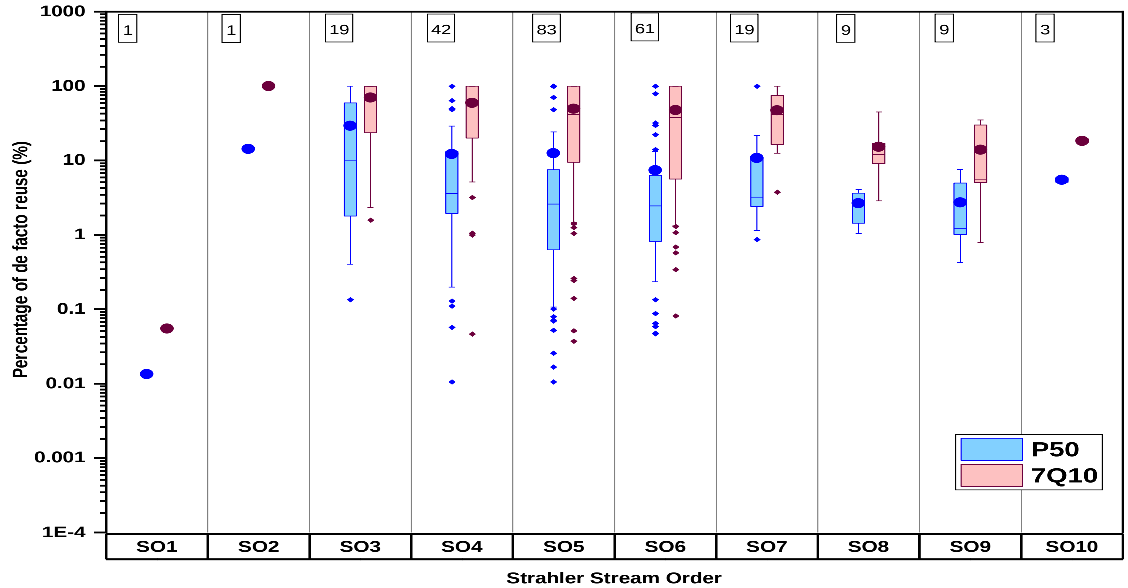


Figure 6.5 Box plot of variation in DFR under 7Q10 low flow and P50 flow condition

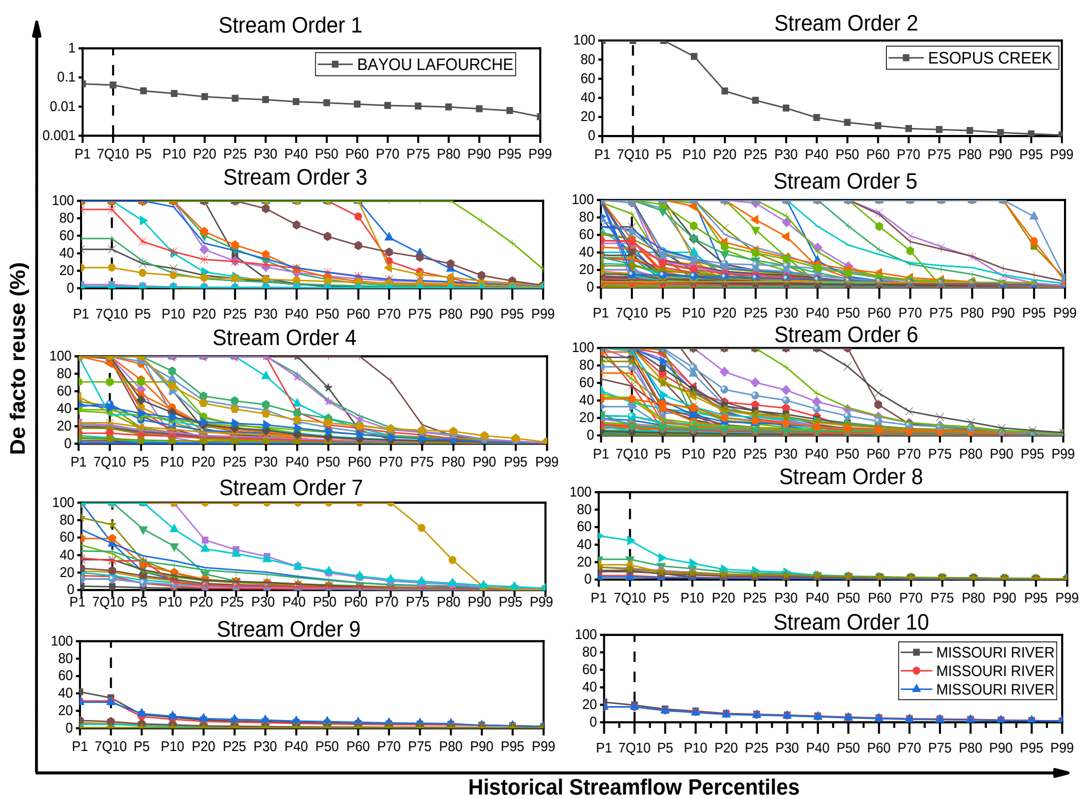


Figure 6.6 Variation in DFR levels as a Function of Historical Streamflow Percentiles Categorized by Strahler Stream Orders

The level of DFR varied substantially to streamflow and Strahler stream order. Historical streamflow percentile indicates the increasing streamflow from P1 (low flow) to P99 percentile (high flow) which results in the decrease in DFR Although high flow condition is likely to have lower DFR due to high dilution factor of the present wastewater in the river, the flood flow can increase watershed connection and transport higher load of contaminants from upstream.

Small rivers are dependent on treated effluent discharge as the variation in the historical streamflow percentile may not alter DFR significantly (Strahler stream order 1 and 2). Higher Strahler stream orders (>8) had a moderate variation in DFR (from 0 to max 40% DFR) while mid-stream orders had a wide variation to historical streamflow percentiles (from 0 to max 100%DFR).

De Facto Reuse at Different Strahler Stream Orders

The distribution of surface water intakes as a function of Strahler stream orders indicates that all DWTPs rely on surface water from local and small streams (Strahler stream orders 1 and 2). Figure 6.7 illustrates that smaller DWTPs had the highest number of surface water intakes on stream order 1 (with 17%) which was doubled in comparison to the number for larger DWTPs (with 7%). Lower-order streams still have sufficient stream flows to meet the low daily water demand of smaller-sized communities while DWTPs serving > 10,000 people tend to extract surface waters from high Strahler stream orders (>8). Overall, as the Strahler stream order increases, there is a decrease in the number of surface water intakes on each stream order.

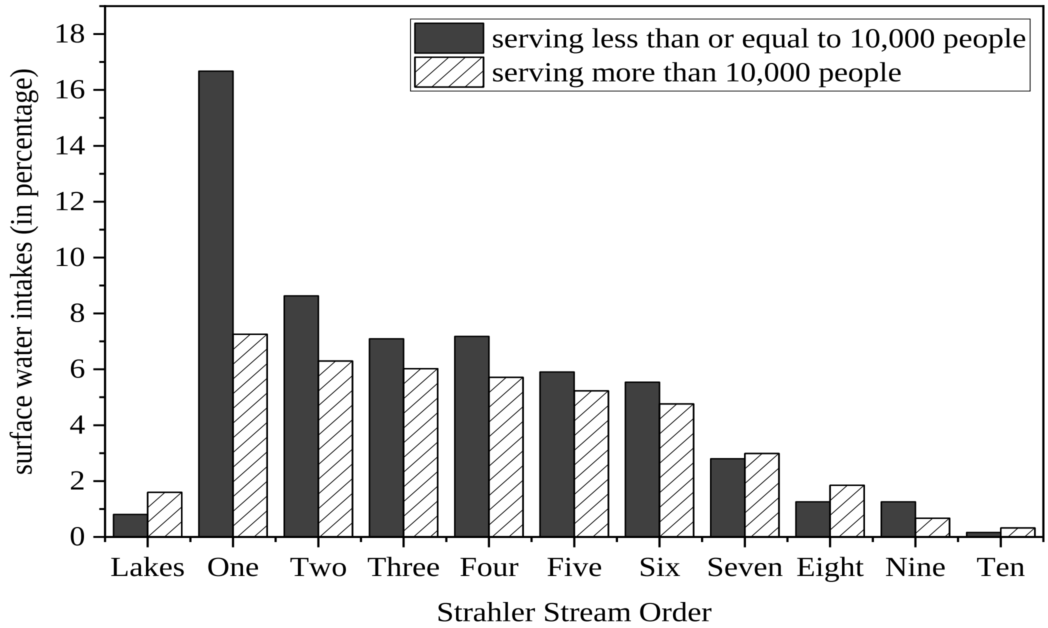


Figure 6.7 Distribution of Surface Water Intakes as a Function of Strahler stream order for Smaller vs Larger DWTPs

Variation in DFR in receiving streams as a function of Strahler stream orders was evaluated in Figure 6.8. Great Lakes had the lowest DFR due to the high dilution factor in lakes. Small stream orders were significantly impacted by treated wastewater effluent discharges as variation in streamflow could not alter much the DFR. Strahler stream orders (3 to 7) had a substantial variation in DFR. DFR can decrease due to the high dilution of increasing streamflow in these streams. High Strahler Stream Order (>8) even had higher mean flow, these big rivers at higher streamflow often dilutes with a large amount of wastewater discharged upstream (Figure 6.8).

Smaller DWTPs extract water most from stream order 1 which are significantly impacted by treated effluent discharges. Using t-test, smaller DWTPs on Strahler Order one was likely to have a significant higher DFR mean than the value for larger DWTPs.

Thus, smaller DWTPs with surface water intakes located on small wastewater-effluent streams were likely to have a higher risk than larger water systems.

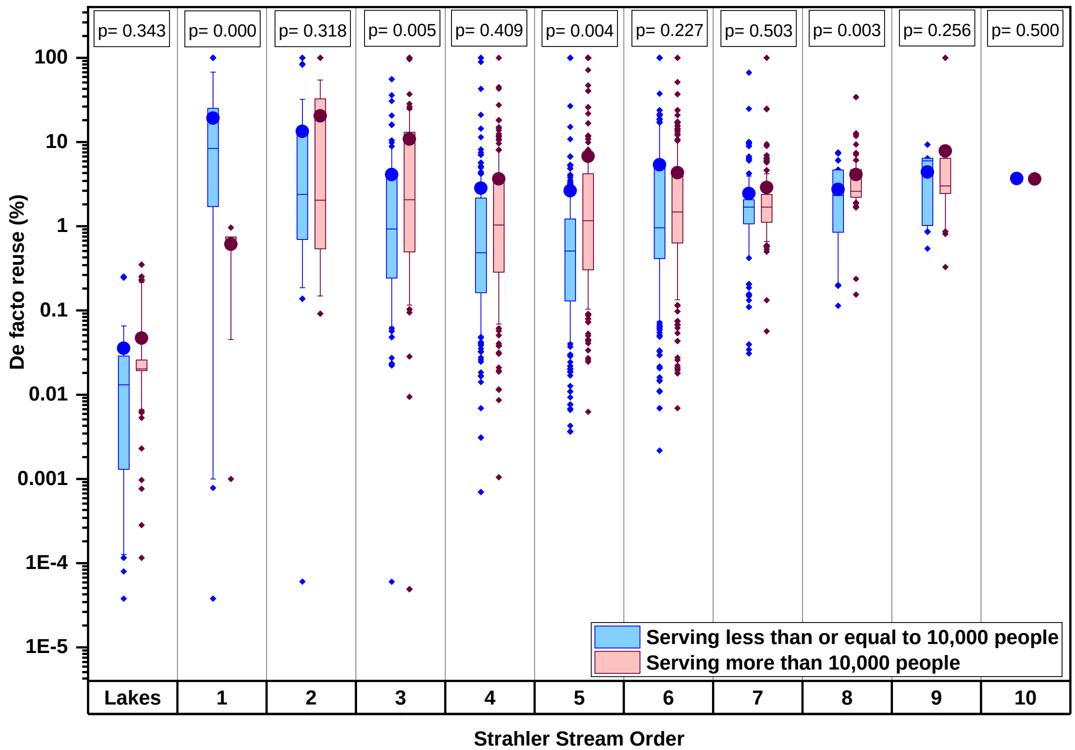


Figure 6.8 Box-and-whisker Plot of Levels of De Facto Reuse Categorized by Strahler Stream Order Under Annual Mean Flow Condition

(Top and bottom of box = 75th and 25th percentiles, respectively; top and bottom of whisker = 90th and 10th percentiles, respectively; line inside box = 50th percentile (median); dot (•) = average)

The two-sample t-test was performed to determine if there is a statistically significant difference between two means of the DFR level in smaller DWTPs and larger DWTPs. A null hypothesis for no difference and the alternative hypothesis is smaller DWTPs have higher mean DFR than larger DWTPs. The significant level alpha is 0.05. There was a significant difference ($p<0.05$) in the mean DFR levels at smaller versus larger DWTPs for Strahler Stream Order 1, 3, 5, and 8. The Cohen's d effect size was also determined and a small effect size is at least 0.20, a medium effect at 0.5, and a large effect size is at least 0.80 (Table 6.1). If changing the significant level (or alpha) from 0.05 to 0.1, the calculated values for p and other descriptive statistic variables for Strahler stream orders have not changed (Table in Supporting Information for alpha = 0.1). Lowering the 95th to 90th confidence interval does not have an impact on the difference in DFR means at smaller versus larger DWTPs.

Table 6.1 Descriptive Statistics in details for t-test with $p < 0.05$

Stream Order	Mean DFR at small DWTPs	Mean DFR at large DWTPs	Standard deviation at small DWTPs	Standard deviation at large DWTPs	Effect size (Cohen's d)	Result of the two-sample t-test, one-tailed	Alternative hypothesis
One	19.2	0.610	28.6	0.304	0.919	$t(44) = 4.36$, $p < 0.05$, $d = 0.919$	Mean DFR at smaller DWTPs > mean DFR at large DWTPs
Three	4.10	10.8	8.99	22.1	0.397	$t(124) = -2.83$, $p < 0.05$, $d = 0.397$	Mean DFR at larger DWTPs > mean DFR at smaller DWTPs
Five	2.6	6.8	12.3	19.6	0.257	$t(454) = -2.92$, $p < 0.05$, $d = 0.257$	Mean DFR at larger DWTPs > mean DFR at smaller DWTPs
Eight	2.74	4.10	2.06	4.41	0.395	$t(189) = -3.01$, $p < 0.05$, $d = 0.395$	Mean DFR at larger DWTPs > mean DFR at smaller DWTPs

The Spearman Rank Correlation Coefficient method was performed using Minitab™ Express software to assess the association between variables (Population served, Stream Orders, and DFR) as stream order is an ordinal variable (First to Tenth) and DFR is a nominal variable (in percentage). The software predicted the Spearman correlation coefficient between each pair of variables. The Spearman correlation coefficient can range in value from -1 to +1. The larger the absolute value of the coefficient, the stronger the relationship between the variables. A value between 0 and 0.3 indicates a weak relationship while a value > 0.7 indicates a strong relationship.

The software results demonstrate a weak positive association between the Population served and Stream Order ($r_s = 0.119$); a strong positive association between the DFR and Stream Order ($r_s = 0.703$); and a weak positive association between the DFR and Population Served ($r_s = 0.103$).

A simple regression analysis using Minitab™ Express software was performed to fit linear or quadratic models with one continuous predictor and one continuous response using least squares estimation. The best-fitting model was selected by the software to test whether there is a correlation between Population served, Stream Order, and DFR at all sized DWTPs. While the software predicted $p < 0.05$, this is not a weak effect of predicting DFR by using the regression model due to a low value of $R^2 = 0.33\%$.

Spatial Distribution of De Facto Reuse in the US EPA Regions

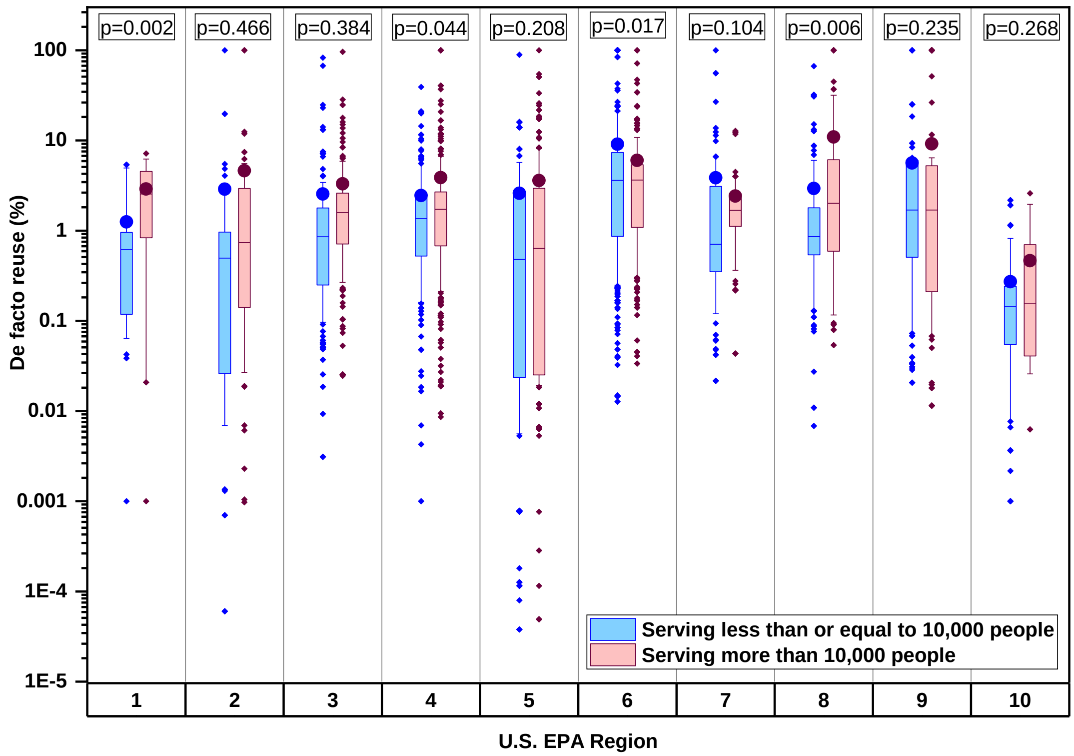


Figure 6.9 Box-and-whisker Plots of Levels of De Facto Reuse Categorized by The EPA Regions under Annual Mean Flow Condition

(Top and bottom of box = 75th and 25th percentiles, respectively; top and bottom of whisker = 90th and 10th percentiles, respectively; line inside box = 50th percentile (median); dot (•) = average)

Smaller DWTPs are likely to have smaller DFR than larger DWTPs on the EPA regions (01, 04 and 08) (t-test, $p < 0.05$). There are ten EPA regions across the country, governing environmental protection programs at states and U.S. Understanding the differences between smaller and larger DWTPs water impacted by DFR can help the EPA region departments to identify, monitor, and prioritize potential environmental public health concerns and opportunities for control actions in their region.

Spatial Distribution of De Facto Reuse in the Climatic Regions

In the United States, the West or the Southwest regions are most impacted by climate droughts. The water availability is limited while the water demand increases due to population growth. The western part of the US observes a reduction in precipitation and snowpack in the mountains. An increase in early snowmelt can lead to a reduction in water availability during summer months to maintain the flow in small streams. This will challenge water managers to provide adequate water in terms of quantity and quality to customers. The Pacific Northwest, northern California, and parts of the southeast has a drier condition (decreasing in streamflow). The findings imply a broad scale capability of the DRINCS model to identify highly impacted areas by de facto reuse at a watershed, a local river, or an EPA region and climatic region. Figure 6.10 illustrates the U.S climatic region (Northeast and Southeast) with ($p < 0.05$) for the two-sample t-test. Smaller DWTPs are likely to have smaller DFR than larger DWTPs in the U.S. climatic regions (Northeast and Southeast).

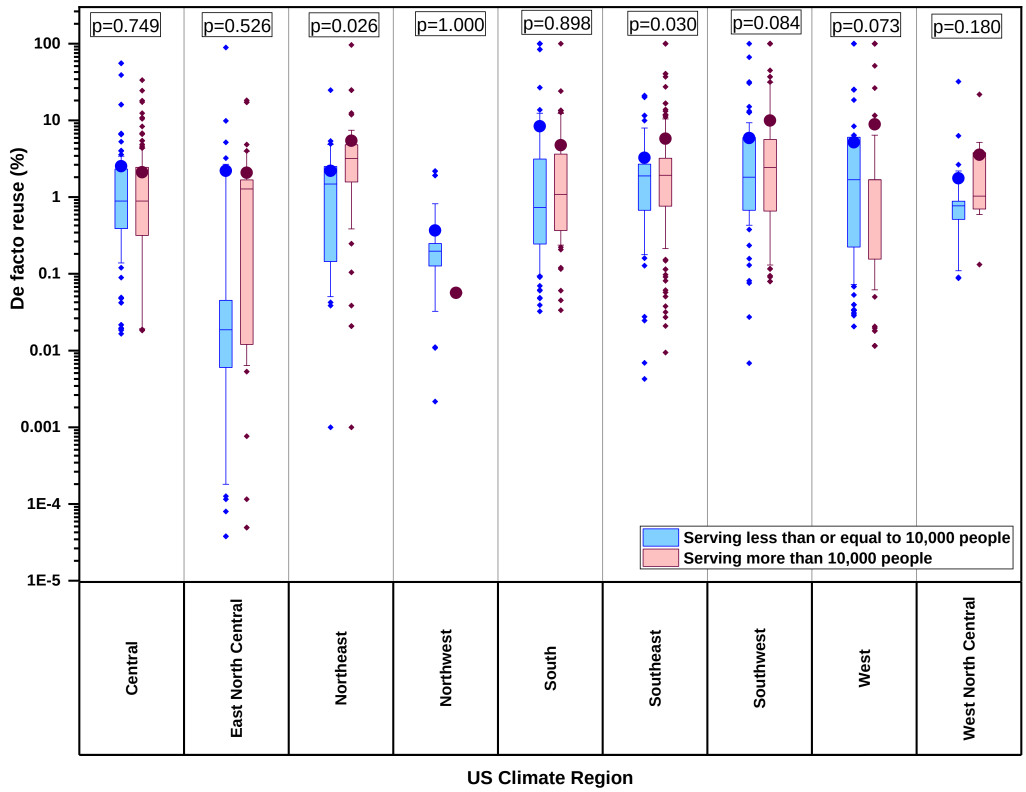


Figure 6.10. Box-and-whisker Plots of Levels of De Facto Reuse Categorized by the US Climatic Regions Under Annual Mean Flow Condition
(Top and bottom of box = 75th and 25th percentiles, respectively; top and bottom of whisker = 90th and 10th percentiles, respectively; line inside box = 50th percentile (median); dot (•) = average)

Installation of Advanced Treatment Technology at Surface Water Systems

Conventional unit processes at WWTPs or DWTPs may not remove many of the trace organic contaminants that may derive from municipal wastewater. An advanced treatment process including microfiltration, ultraviolet advanced oxidation process, ozone, biological activated carbon, membrane bioreactors, or nanofiltration. Advanced oxidation process includes ozonation, and/or combined with hydrogen peroxide, and UV light and/or combined with hydrogen peroxide (UV/H_2O_2) to produce hydroxyl radicals, then effectively oxidize contaminant of emerging concern (CECs).

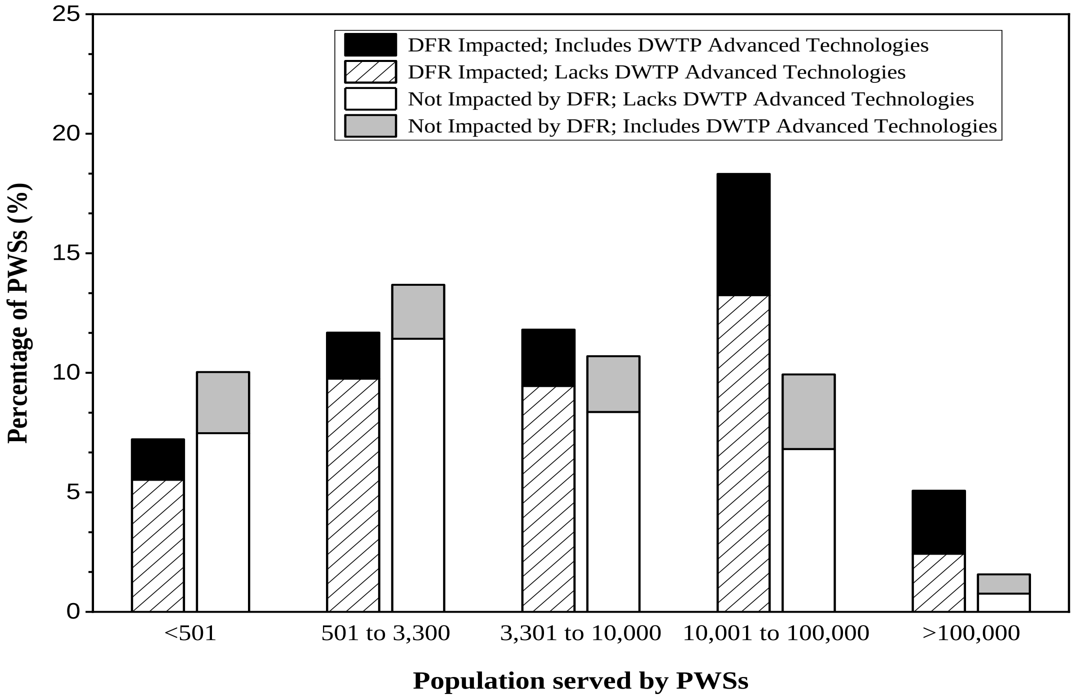


Figure 6.11 Distribution of DWTPs Employed with Advanced Technologies and level of DFR

In total, 25% of all the DWTPs have installed advanced unit processes. The number of smaller DWTPs employed advanced unit process was slightly higher than larger water systems with 13.1% and 12.6%, respectively. Meanwhile, the number of smaller DWTPs without advanced unit processes was doubled than the larger DWTPs with 52.1% and 23%, respectively (Figure 6.11).

Population Exposed to De Facto Reuse in Surface Water

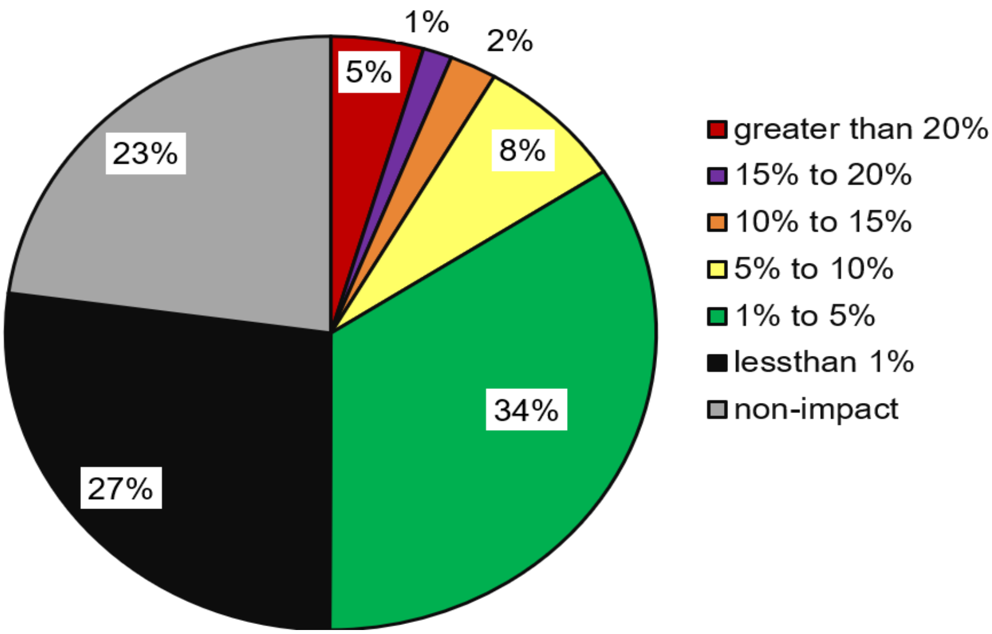


Figure 6.12 Distribution of Total Population Served by DWTPs Categorized by Levels of De Facto Reuse

The total population served of DWTPs impacted DFR at each category of DFR level was used to estimate potential risks associated with de facto reuse. The highest total of the population served using surface water (34%) with DFR range between 1% and 5% (Figure 6.12). DWTPs serving 33.43 million people (of the 146.34 million people in total) were not impacted by DFR. More than 73 million people relied upon source water with a wastewater content of DFR <1% under annual mean flow conditions and 12.3 million people relied on source water with a wastewater content of DFR >10% during annual mean flow conditions (Figure 6.13). Population served by larger systems tend to have a higher population potentially exposed to de facto reuse in surface water than smaller systems as they supply water to a larger size of customers.

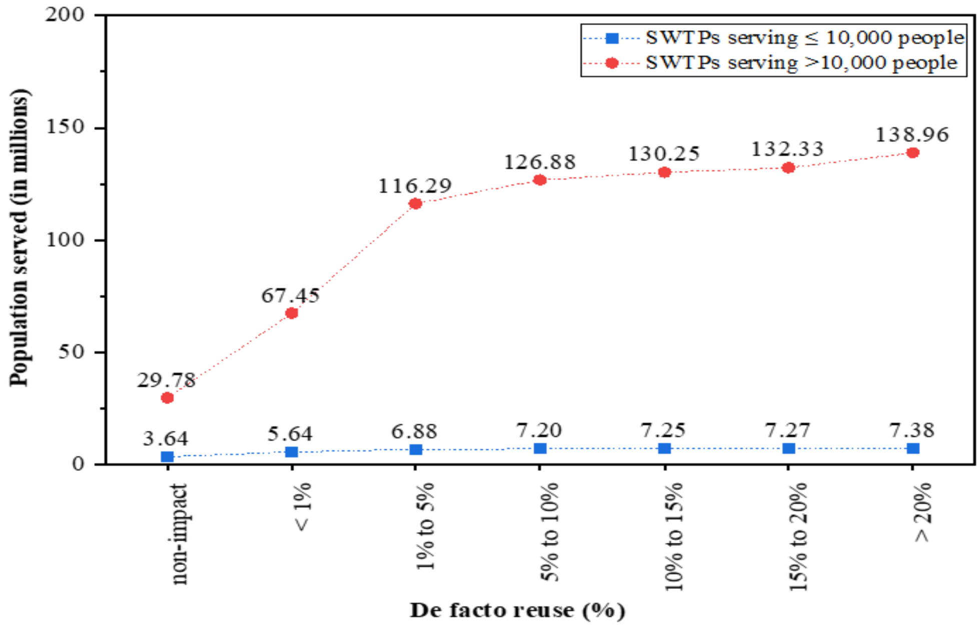


Figure 6.13. Cumulative of Total Population Served of DWTPs Impacted by DFR, Grouped by DFR Level, and Categorized by DWTP Sizes

As large WTPs serve populated communities, large WTPs are likely to have a higher exposed population to the risks of CECs in surface water than the smaller WTPs. The DRINCS model predicted, at DFR >1%, the larger DWTPs could pose risks of CECs to a population as 40 times as those served by smaller systems with 71 million and 1.7 million people, respectively. The total exposed population to the risks of CECs to source water was estimated at 73 million (at DFR>1%) and 12.3 million people (at DFR>10%) (Figure 6.13).

Conclusion

Deterioration in the quality of surface water can pose health risks to human consumption. The provision of safe surface water sources is a major challenge, especially in small water systems that often lack the technical, financial, and human resources for proper and efficient operations. The DRINCS model was expanded to include smaller

DWTPs (N = 6,045 SW intakes at 3,984 DWTPs) in the U.S. The preliminary results for Texas predicted that two-thirds of all surface water (SW) intakes were impacted by at least one WWTP upstream. The level of DFR at SW intakes in Texas ranged between 1 to 20% under average streamflow and exceeded 90% during mild droughts. Smaller DWTPs in Texas had a higher frequency of DFR than larger systems while < 10% of these DWTPs employed advanced technologies capable of removing CECs. Nationally, > 40% of SW intakes at all sized DWTPs across the U.S. were impacted by DFR under average flow (N = 2,917 of 6,826). Smaller DWTPs had a higher frequency of DFR than larger DWTPs with 1,504 and 1,413, respectively. The research hypothesis was rejected as there was likely no statistical difference between the levels of DFR in surface water for smaller versus larger DWTPs (t-test, $t(2917) = -0.536$, $p = 0.274$). Smaller DWTP intakes located on lower Strahler stream orders were predicted to have a higher frequency of DFR than those for larger water systems. Low order streams are frequently dependent on treated wastewater effluents and vulnerable to high variation in the level of de facto reuse under mild droughts (7Q10 or Q95 low flow). Smaller DWTPs are often supplied by lower stream orders (1 and 2) while DWTPs serving > 10,000 people require larger and more consistent river flowrates (3, 5, and 8) to sufficiently provide water to the public.The study investigated the current employment of advanced unit processes (i.e., activated carbon, ozonation, ultraviolet, nanofiltration) at DWTPs to evaluate the capability of removing CECs from source water. Smaller DWTPs lack a doubled number of advanced technologies compared with larger systems. There is no installation of any advanced unit processes at 52% and 23% for smaller versus larger DWTPs, respectively. Thus, when de facto reuse occurs, plus the lack of advanced technology at DWTPs can pose higher risks to CECs to the communities they serve. As larger DWTPs frequently supply surface water to populated communities, the total exposed population to the risks of CECs in source water could be extremely significant at larger DWTPs than smaller systems. The model estimated at DFR >1%, this could pose risks to a population served by larger DWTPs as 40 times as those served by smaller systems with 71 million and 1.7 million people, respectively. The model estimated a total of 73 million people relying on surface water impacted by DFR >1%. When predicted DFR was > 10%, the total exposed population to the risks of CECs to source water was estimated as 12.3 million people. Future studies can use DFR results to conduct epidemiological and risk assessment studies for communities impacted by wastewater derived CECs and identify communities that would benefit from advanced treatment processes that remove CECs. To improve water quality and achieve sustainable service, decision-makers and the population they represent must be notified of the relevant health issues concerning water and may be aware of the occurrence of de facto reuse in surface water supplies. As investment in advanced treatment technology may be costly and slow in coming for small community water systems impacted by DFR, it is possible to consider some quick and cost-effective solutions such as properly managed decentralized water production (e.g., point-of-entry, point-of-use treatment unit, and/or bottled water). Advanced treatment installed at DWTPs and increased monitoring of small Strahler stream orders (Order 1 and 2) associated with the highly impacted by DFR could be considered within the SDWA to protect the source water quality and public health. The adjustment could consider that smaller utilities may be impacted more due to financial trains or may have fewer resources to install advanced treatment. There is a need for a warrant policy for smaller utilities such as lower standards to control source water or supporting grants to upgrade the water plants. Findings from the DRINCS model could facilitate the development of contamination prediction tools or research on monitoring of CECs in surface water to understand and ensure clean and safe water.

Acknowledgments

This research was partially supported by the National Science Foundation (EEC-1449500, SES-1462086, CBET-1804229, and DEB-1637590), the Vietnam Education Foundation (VEF), Decision Center for a Desert City (DCDC), and ASU's Future H2O initiative. We would like to thank two VEF Fellow (Khoi Nguyen and Hai Nguyen) for help with Python programming and Laurel Passantino for technical editing.

Supporting Information

Table S 1 The U.S. Community Water Systems: Water-type and population served (SDWIS, the first quarter of the year 2020)

Groundwater or Surface Water Type	Number of Community Water Systems	Total Population Served
Groundwater	37,975 (76.6%)	90,203,324 (29%)
Surface water	11,600 (23.4%)	220,942,538 (71%)
Total	49,575	311,145,862

The Safe Drinking Water Act (SDWA) passed by Congress in 1974 is responsible for regulating contaminants and drinking water quality at more than 150,000 public water systems which serve about 95% of the United States population (or more than 300 million people).

Table S 2. The U.S. Community Water Systems using Surface Water: Size distribution and population served (data from the DRINCS model)

Size of Public Water Systems	Population served	Number of Community Water Systems Serving This Size Community	Total Population Served by Systems This Size
Very Small	Under 500	681 (17.3%)	113,715 (0.1%)
Small	501 – 3,300	1,001 (25.4%)	1,775,342 (1.2%)
Medium	3,301 – 10,000	888 (22.5%)	5,486,333 (3.7%)
Large	10,001 – 100,000	1,115 (28.2%)	36,014,505 (24.6%)
Very Large	More than 100,000	262 (6.6%)	102,949,105 (70.3%)
	Total	3,947	146,339,000

8% of U.S. community water systems provide surface water to 82% of the U.S. population through large municipal water systems.

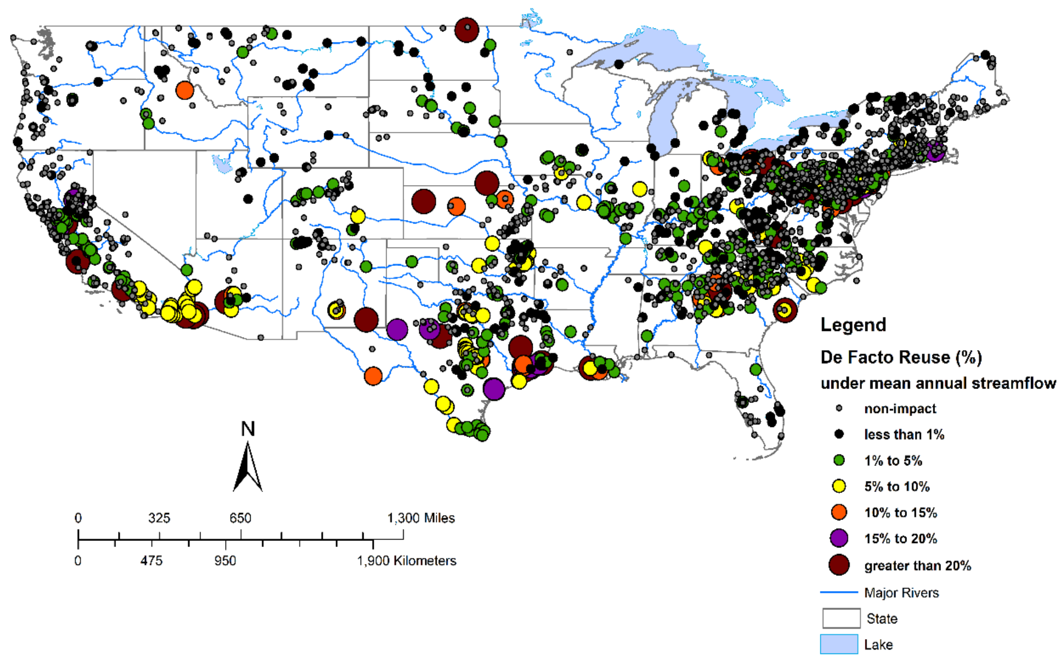


Figure S 1. Geographical Distribution of De Facto Reuse Present at Inactive Surface Water Intakes in the U.S

The full DRINCS dataset contains 9,702 surface water intakes at 5,575 DWTPs including infiltration gallery sites at groundwater under the direct influence of surface water. This map shows 2,876 surface water withdrawal sites that are inactive.

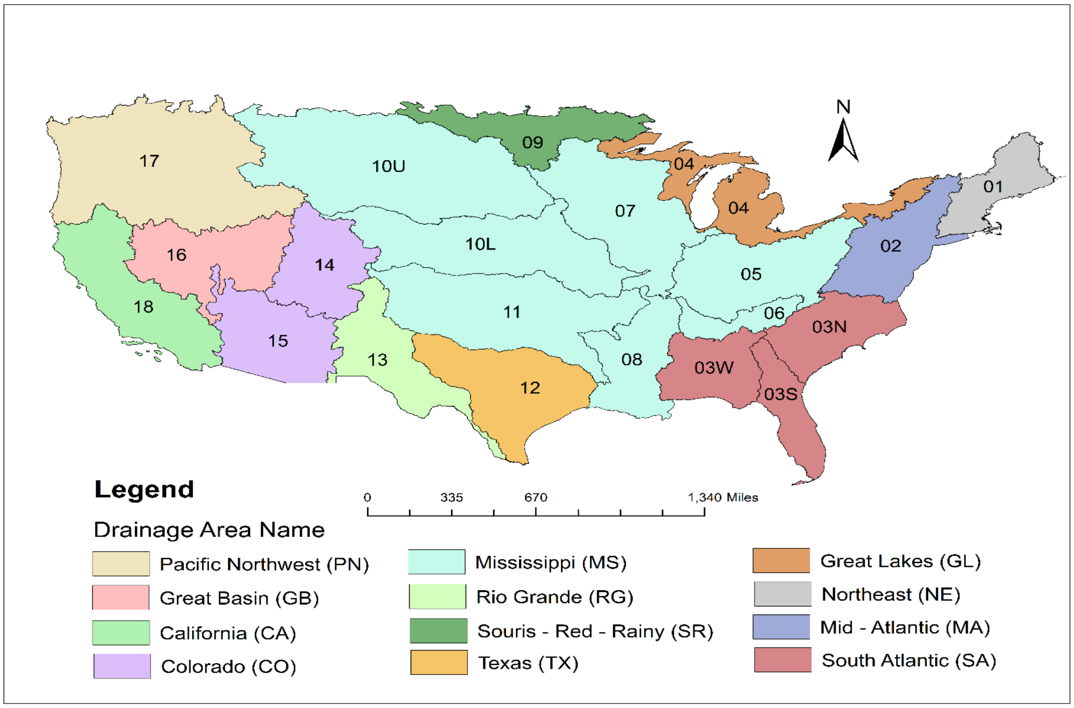


Figure S 2. Drainage Area Name Associated with Vector Processing Units in the Contiguous United States

In NHDPlus V2, the processing units are referred to as Vector Processing Units (VPU) in the continental United States for vector data which includes 12 VPUs across the country. Another term is the Hydrologic Region for vector data which includes 18 Hydrological Unit Code (HUCs) for the conterminous U.S. Some VPUs share the same hydrologic regions such as Northeast VPU for region 01, Mid Atlantic VPU for region 02, Great Lakes VPU for region 04, Souris – Red – Rainy VPU for region 09, Texas VPU for region 12, Rio Grande VPU for region 13, Great Basin for region 16, Pacific Northwest for region 17 and California VPU for region 18. This is not the case for several VPUs which integrate several HUCs such as the South Atlantic VPU combining hydrologic region 3S, 3N, and 3W or the Mississippi VPU contains hydrologic region 05, 06, 07,08, 10U, 10L, 11 and Colorado VPU contains hydrologic region 14 and 15.

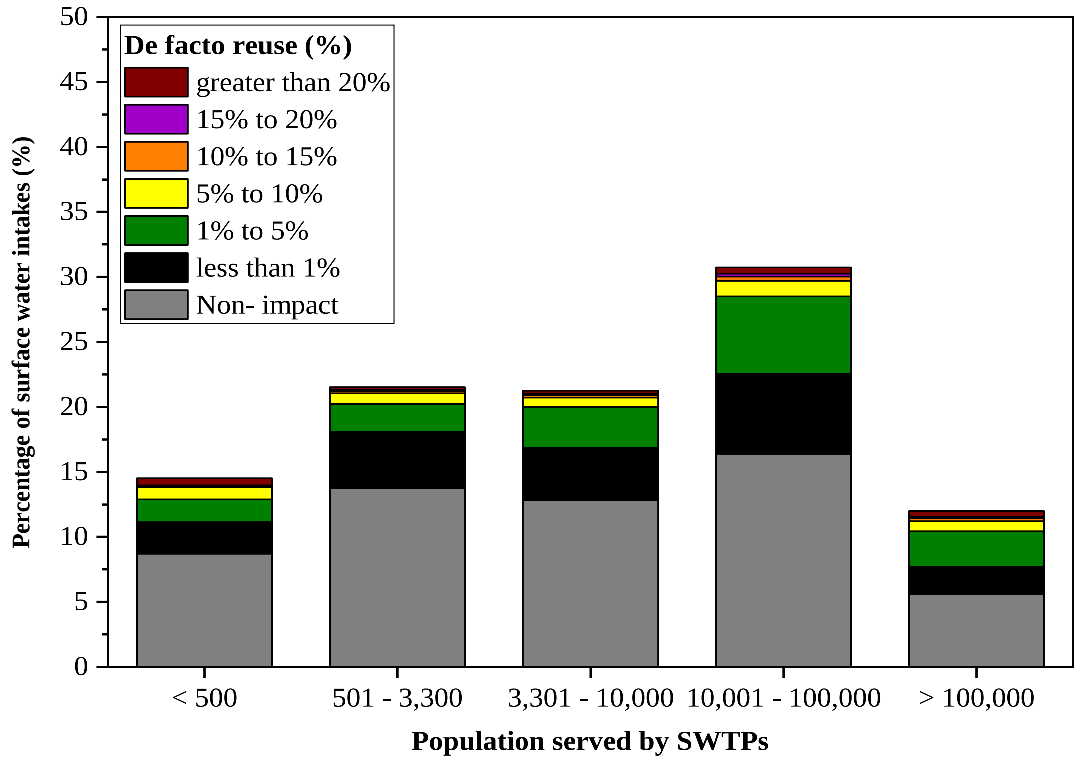


Figure S 3. Distribution of The Number of Surface Water Intakes Impacted by Population Served as a Function of DFR Levels

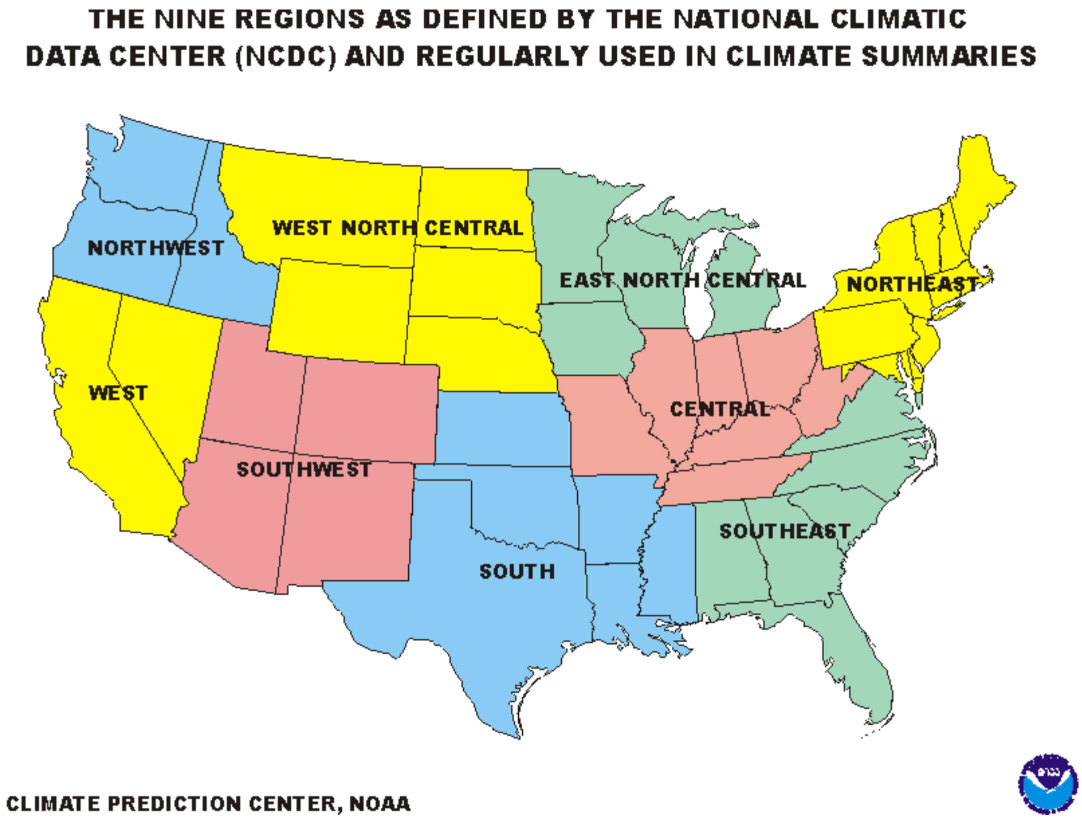


Figure S 4. Climatic Regions in the United States

The U.S climatic conditions vary by region. The **Northeast** is characterized by a fairly diverse climate, with bitterly cold winters and semi-humid summers, especially to the south. On the **West Coast,** it is expected to have cool, wet winters and dry, cool summers. Meanwhile, the **Southeast** has a humid and sub-tropical climate, with warmish winters hot summers. The **Midwest** is similar in that summers are humid, although winters are usually much colder than in the Southeast.

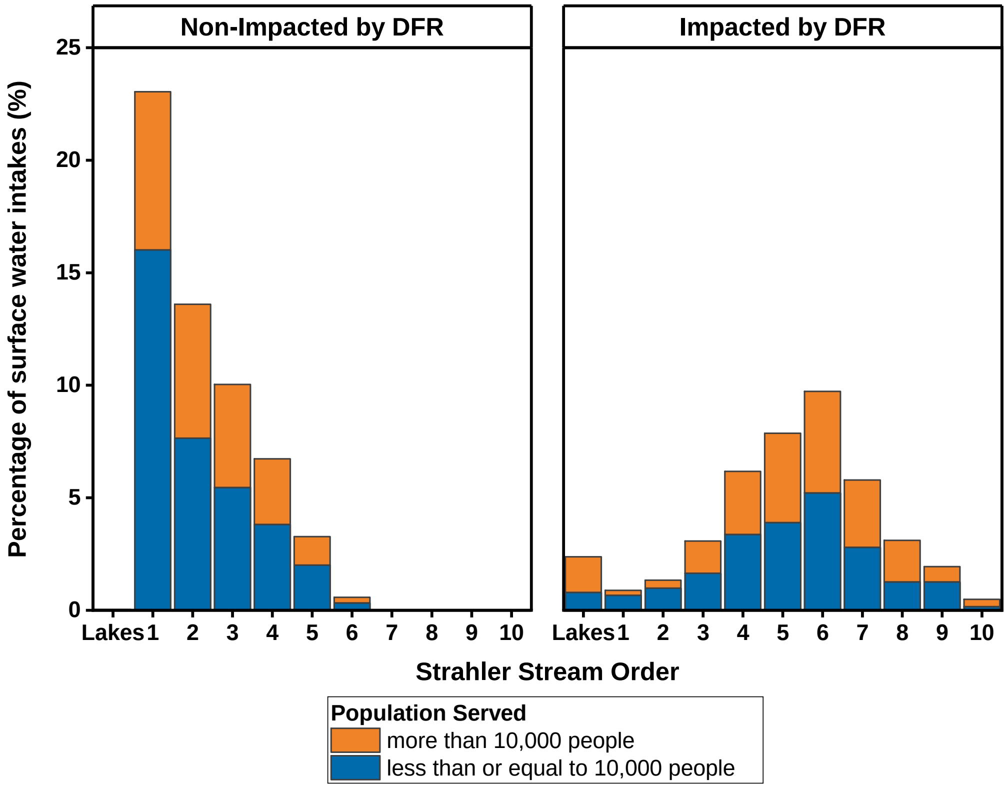


Figure S 5. Number of Surface Water Intakes Impacted or Not Impacted in Two groups, Larger Versus Smaller DWTPs as a Function of Strahler Stream Order

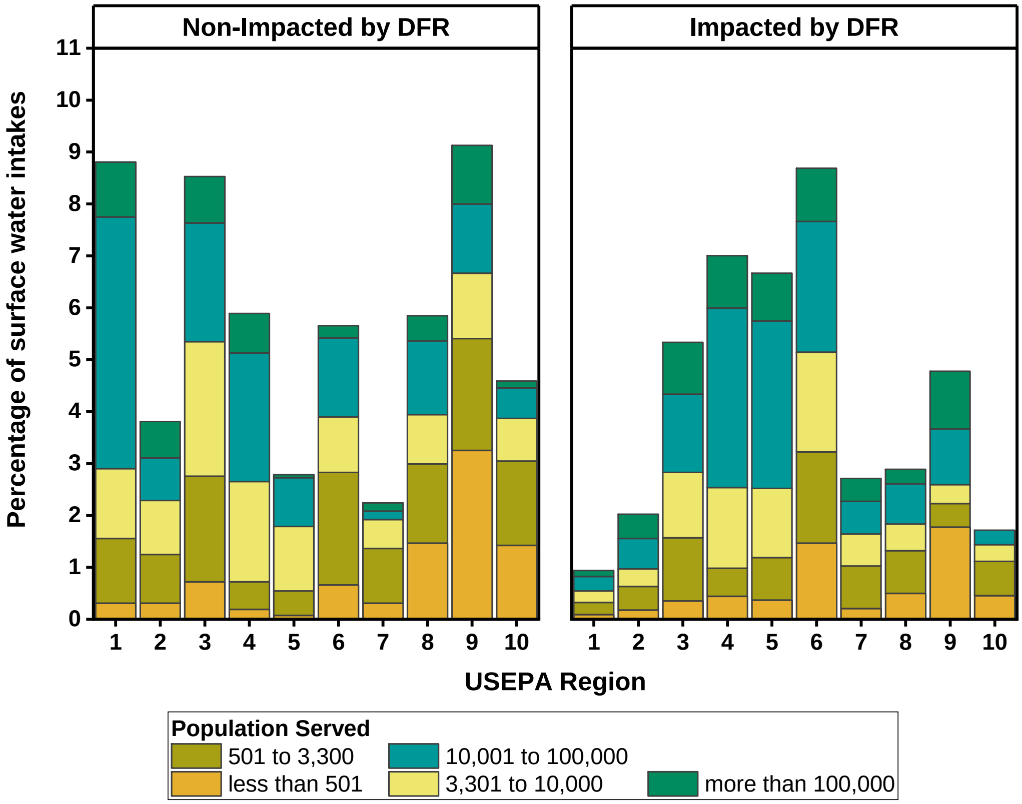


Figure S 6. Distribution of DFR by different sized DWTPs grouped by the USEPA Regions

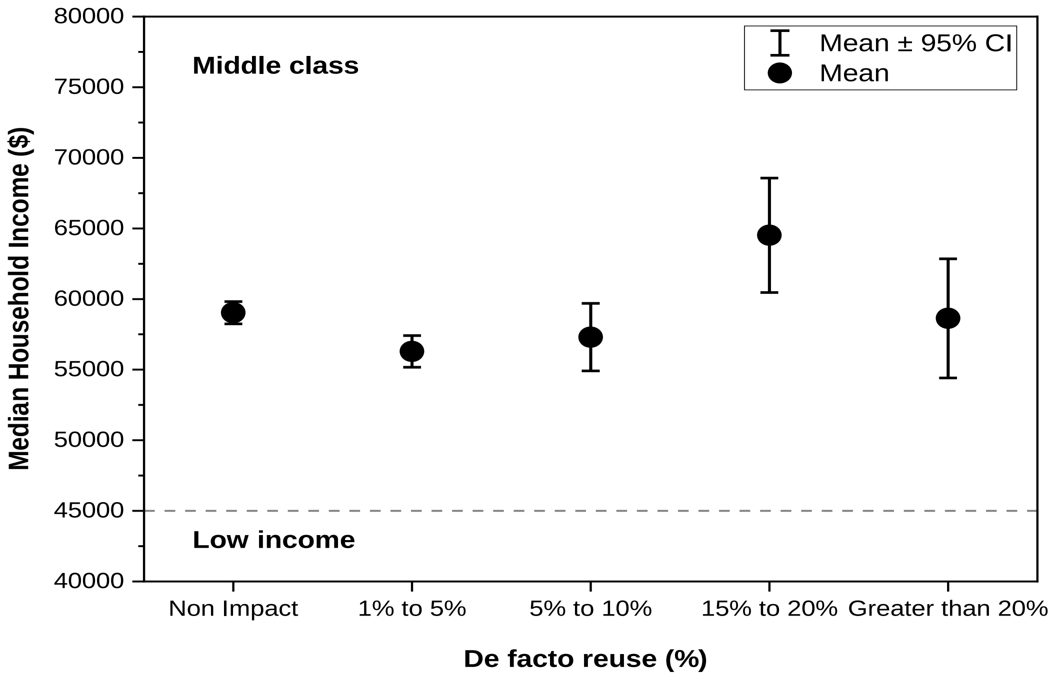


Figure S 7. Mean Household Income Versus De Facto Reuse

Each DWTP is in a ZIP/FIPS code. The mean household income is reported in a ZIP/FIPS code. Surface water supply to the living area of the residences who have middle class means household income. The level of DFR various between 40,000 and 65,000 dollars of median household income.

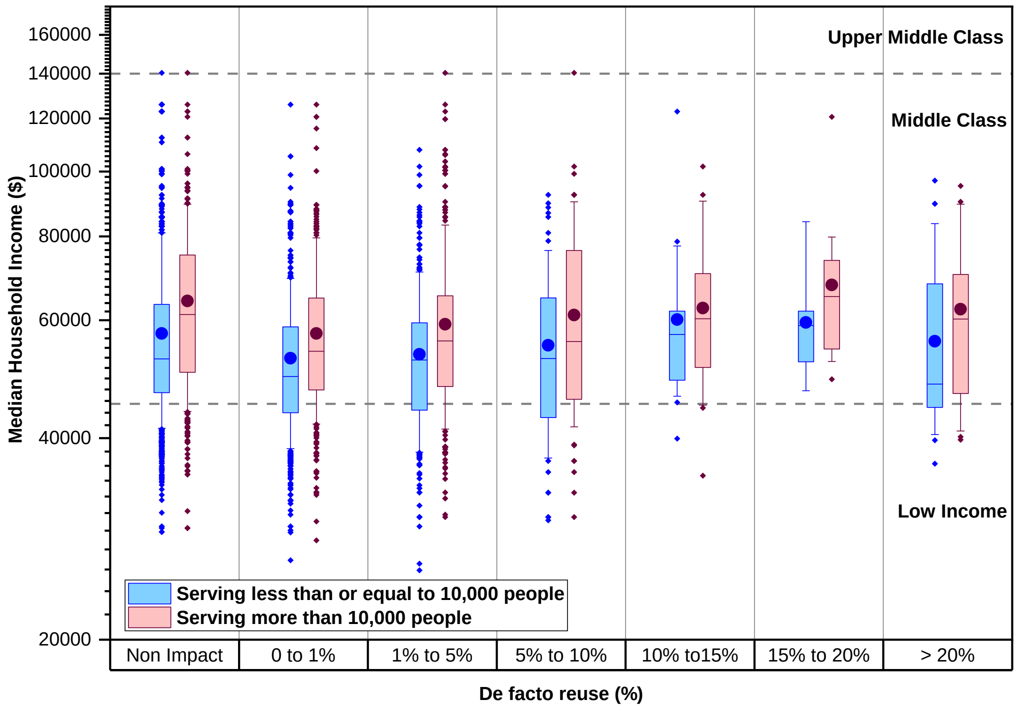


Figure S 8. Mean Household Income Versus De Facto Reuse Categorized by DWTP sizes are grouped by DFR levels

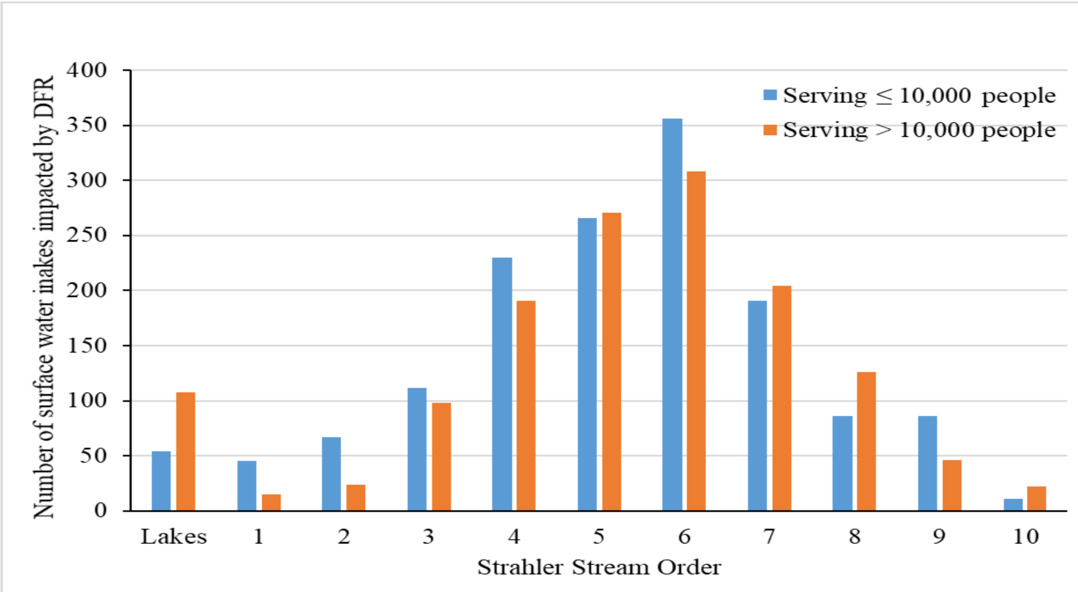


Figure S 9. Distribution of Surface Water Intake Impacted by DFR as a function of Strahler Stream Order

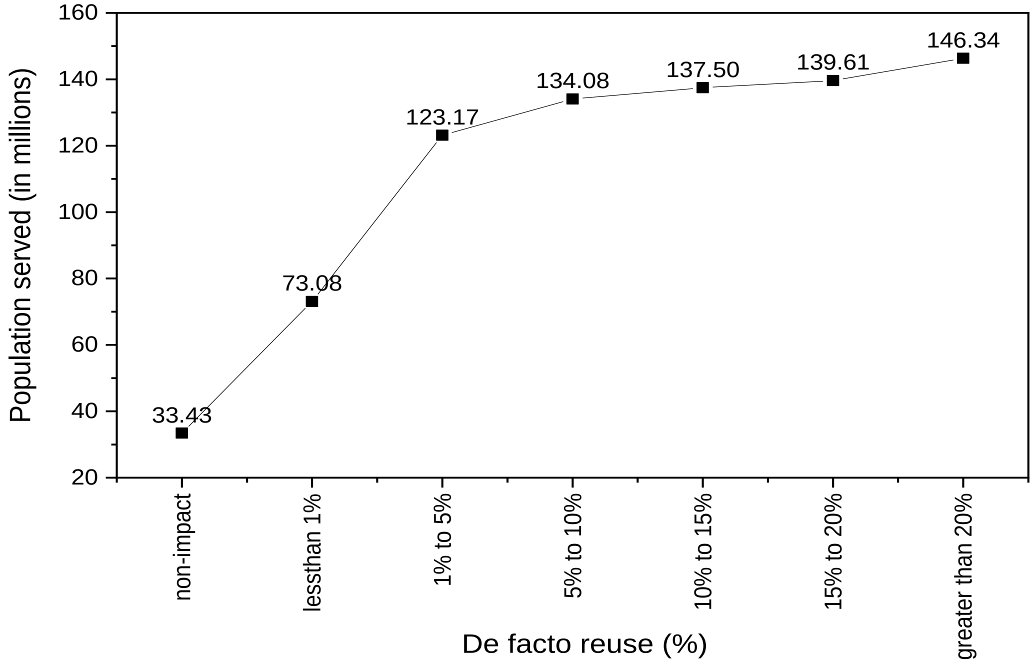


Figure S 10. Cumulative of Number of DWTPs Impacted by DFR, Grouped by DFR Level and Categorized by DWTP Size

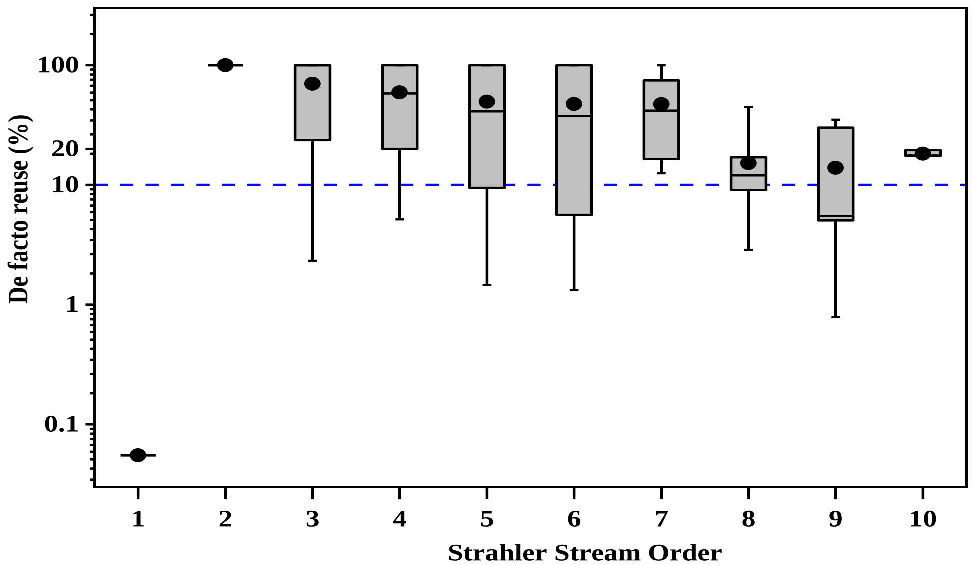


Figure S 11 Box-and-whisker Plots showing Variation in Levels of De Facto Reuse at Strahler Stream Order under 7Q10 Low Flow Condition, classified based upon Strahler Stream Order

Figure S 11 illustrates DFR under 7Q10 low flow condition at all 274 stream gauges which most of them had a mean DFR level (> 50%). More available stream gauges are located on higher Strahler stream order while there is only one station on stream order 1 and 2.

Table SI 3. Descriptive Statistics for alpha 0.1 at all Strahler Stream Orders

	Alpha = 0.1								
Alternative Hypothesis	Does not equal to			Small > Large			Small < Large		
Strahler Stream Order	90% Confidence Interval	t	p	90% Lower Bound for Difference	t	p	90% Upper Bound for Difference	t	p
Great Lakes	(-0.0312, 0.0085)	-0.95	0.343	-0.0268	-0.95	0.828	0.004	-0.95	0.172
One	(11.46, 25.81)	4.36	0	13.08	4.36	0	24.19	4.36	1
Two	(-18.92, 4.74)	-1.01	0.318	-16.24	-1.01	0.841	2.05	-1.01	0.159
Three	(-10.70, -2.79)	-2.83	0.005	-9.82	-2.83	0.997	-3.67	-2.83	0.003
Four	(-2.468, 0.819)	-0.83	0.409	-2.105	-0.83	0.796	0.455	-0.83	0.204
Five	(-6.44, -1.80)	-2.92	0.004	-5.92	-2.92	0.998	-2.31	-2.92	0.002
Six	(-0.386, 2.518)	1.21	0.227	-0.065	1.21	0.114	2.196	1.21	0.886
Seven	(-1.510, 0.637)	-0.67	0.503	-1.273	-0.67	0.749	0.399	-0.67	0.251
Eight	(-2.103, -0.612)	-3.01	0.003	-1.938	-3.01	0.999	-0.777	-3.01	0.001
Nine	(-8.38, 1.56)	-1.15	0.256	-7.26	-1.15	0.872	0.44	-1.15	0.128
Ten	(-0.0578, 0.1351)	0.68	0.5	-0.0357	0.68	0.25	0.1131	0.68	0.75

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CHAPTER 7

DISSERTATION SUMMARY AND FUTURE RESEARCH DIRECTIONS

Summary

The overall goal of this dissertation is to quantify levels of de facto reuse at surface water intakes for smaller DWTPs (serving ≤ 10,000 people) across the United States by expanding the De facto Reuse Incidence in our Nations Consumptive Supply (DRINCS) model and develop a programmed ArcGIS model tool capable of proximity analysis between upstream WWTPs and DWTPs. The hypothesis tested whether smaller surface water DWTPs (serving ≤ 10,000 people) were likely to be more impacted by DFR than larger systems (serving > 10,000 people). The research in this dissertation fulfilled the dissertation objectives which as following:

1) Demonstrated how predicted DFR by the DRINCS model can be confirmed with field observation of CECs occurrence at DWTPs

2) Fully ground-truthed location of all surface water intakes at DWTPs (N = 9,702) and wastewater effluent outfalls at WWTPs (N = 16,161) in the continental U.S

3) Quantified and compared the level of de facto reuse for smaller versus larger public water systems across the United States by expanding a previous version of the DRINCS model

4) Developed an automated proximity tool to determine the travel times between multiple WWTPs and downstream DWTP

5) Advanced the model with an automation process (the DRINCS version 2.0)

A summary of the key findings for each dissertation chapter (from Chapter 3 to Chapter 6) was provided in this Chapter.

Key Findings

Chapter 3: Nguyen, T., Westerhoff, P., Furlong, E.T., Kolpin, D.W., Batt, A.L., Mash, H.E., Schenck, K.M., Boone, J.S., Rice, J. and Glassmeyer, S.T., 2018. Modeled de facto reuse and contaminants of emerging concern in drinking water source waters. Journal-American Water Works Association, 110(4), pp. E2-E18.

- Demonstrate how predicted DFR by the DRINCS model can be confirmed with field observations of CECs occurrence at DWTPs
- Compare specific sampling effort and predicted DRINCS results
    - Analyzed 192 organic CECs at surface water intakes from 22 DWTPs using surface water across the U.S
    - Predicted DFR at 22 of 25 DWTP surface water public water systems across the U.S (3 groundwater DWTPs)
- Predict 19 of the 22 DWTPs with at least one upstream WWTP discharge
    - Level of DFR range from 0 to 12.8% under annual mean streamflow
- Evaluate impacts of varying streamflow (daily, seasonal, and annually) on level of DFR
    - Higher Strahler stream orders have lower DFR values
    - SW intakes on fifth- and sixth-order streams had the highest DFR values
    - 84% is the highest DFR under low flow conditions (Q95)
- Confirm the observed correlation between chemicals detected and DRINCS modeling results (midsize water bodies)
- Evaluate the presence of upstream WWTPs (number and design capacity) to surface water sources at 19 of 22 DWTPs across the U.S

    - Between 0 to > 1,000 WWTPs upstream DWTP sources
    - Proximal distance between WWTPs and DWTPs within 10 miles or hundreds of miles upstream
    - 82% (or 3,615) of total upstream WWTPs are small design capacity (< 1mgd)

Chapter 4: DRINCS version 2.0 was upgraded by implementing a three-step approach. First, the previous DRINCS was expanded from only larger DWTPs serving > 10,000 people to include all SW intakes in the continental USA and updated with the most recent database for DWTPs and WWTPs. Secondly, all the locations of WWTP outfalls to surface water and DWTP surface water intakes in the updated database were fully ground-truthed to improve the accuracy of the DRINCS. The final step was to automate the conceptual model for proximity analysis within the DRINCS model.

- Update the DRINCS with a fully ground-truthed geospatial database for DWTPs (N = 9,702) and WWTPs (N =16,161), and the NHDPlus Attribute of streamflow data
- Create four Python stand-alone script tools and integrate them into an automated proximity tool within the ArcGIS™ framework

Chapter 5: Nguyen, T.T., and Westerhoff, P.K., 2019. Drinking water vulnerability in less-populated communities in Texas to wastewater-derived contaminants. *npj Clean Water*, 2(1), pp.1-9.

- Expand the DRINCS model to include an additional number of smaller DWTPs serving ≤ 10,000 people (325 surface water intakes at 245 smaller DWTPs) into 270 SW intakes at 155 larger DWTPs

- Predict two-thirds of all SW intakes in Texas were impacted by DFR
- Under average streamflow DFR level range between 1 to 20% and exceed 90% during mild droughts
- Smaller DWTPs have a higher number of facilities impacted by DFR than larger systems with 46% and 30%, respectively
- Fewer than 10% of smaller DWTPs in Texas employed advanced technologies capable of removing CECs (compared with ~ 30% at larger DWTPs)
- A pilot study in the Trinity River basin with 151 WWTPs and 45 DWTPs
    - Proximal distances are analyzed for selected 10 DWTP intakes
    - Nine DWTPs have 20 to 30 WWTPs within 10-25 mile upstream
    - Most WWTPs are located 100-300 miles upstream of DWTP intakes
    - Accumulated WW range from <10 MGD (within 10 miles) to nearly 1,400 MGD (>1,000 miles)
    - For a 0.328 ft/sec streamflow, ~50% of ibuprofen degraded within 60 miles

Chapter 6: In preparation for peer-reviewed submission to *Environmental Health Journal*: Nguyen, T., Westerhoff, P., Acero, J. Unplanned Water Reuse Impacts on Drinking Water for Smaller-versus Larger Systems across the United States.

- Quantify the extent of de facto reuse in surface water for smaller DWTPs (serving ≤ 10,000 people) in the continental U.S
- Compare the impacts of de facto reuse in surface water for smaller DWTPs (3,911 surface water intakes at 2,570 DWTPs) versus large water systems (2,915 intakes at 1,377 DWTPs)

- Smaller DWTPs had a higher frequency of DFR than larger water systems with 1,504 and 1,413 respectively
- The difference between the level of DFR at smaller versus larger DWTPs were statistically unclear (t-test, $p > 0.05$)
    - Smaller DWTPs were likely to have a statistically higher level of DFR at SW intakes located on Strahler Stream Order 1
    - Larger DWTPs tend to have higher DFR on Strahler stream order 3, 5 and 8
- Smaller communities relied on SW from a low order stream impacted by DFR together with lacking a doubled number of advanced DWTP unit processes than larger systems could have high risks to CECs
- Levels of DFR for larger DWTPs were statistically higher on mid-size stream orders than those for smaller systems
- As they serve large communities, this could pose risks to a population as 40 times as those served by smaller systems
- The total exposed population to the risks of CECs to source water was estimated at 73 million (at DFR>1%) and 12.3 million people (at DFR>10%)
- Future studies can use DFR results to conduct epidemiological and risk assessment studies for communities impacted by CECs and identify communities that would benefit from advanced treatment processes that remove CECs.

Conclusions

The research in this dissertation has completed a nationwide assessment of the extent of de facto reuse in surface water sources in the continental U.S by expanding from only larger DWTP intakes (serving > 10,000 people) to include smaller DWTP intakes (serving > 10,000 people). The DRINCS model was successfully upgraded to the DRINCS version 2.0. The updated model provides national data on over 16,000 wastewater effluent outfalls from WWTPs and nearly 10,000 surface water intakes for all sized DWTPs with a fully ground-truthed geospatial database. The DRINCS model has been confirmed its validity by a qualitative comparison between CECs in grab samples of source water and the modeled DFR at 22 DWTPs using surface water across the U.S. This confirmation facilitates the utility of DRINCS model to identify DWTPs at higher risks for CECs occurrence, treatment technology testing, and future sampling and monitoring.

The DRINCS has been upgraded successfully into version 2.0 with an automation process and a programmed GIS model tool to determine the proximity analysis between upstream WWTPs and DWTPs. The new functionality of the DRINCS model has been developed to perform geoprocessing analysis. At first, a conceptual model of proximity analysis between upstream WWTPs and DWTPs was created. Automated proximity analysis was developed to estimate travel time during which pollutant transformation can occur between upstream WWTPs and DWTPs. The calculated proximity indicators were used as a secondary index of relative risks associated with CECs from individual WWTPs that slowly degrade in the environment. Four stand-alone Python scripts were written and integrated into a ModelBuilder model tool within the ArcGIS™ framework to automate the tracing upstream process and determine the proximity index. A dialog box of the tool provides an easy-to-use interface to the user and it is easier to run and share the model tool with others.

A preliminary study has been done for Texas to extend the previous DRINCS model to include all sized DWTPs using surface water and compare the level of DFRs at smaller (serving ≤ 10,000 people) versus larger (serving > 10,000 people). The model predicted that two-thirds of all surface water (SW) intakes were impacted by at least one WWTP upstream. Smaller DWTPs in Texas had a higher frequency of DFR than larger systems while fewer than 10% of these DWTPs employed advanced technologies capable of removing CECs. The proximal distances between 151 WWTPs and 45 DWTPs determined in a pilot study in the Trinity River basin to examine wastewater surrogates from upstream WWTP outfalls to surface water source (e.g., rivers, lakes, canals, reservoirs) and their transformation due to natural attenuation in surface water (e.g., decay, degrade, loss or transformed products).

Nationally, the upgraded DRINCS model version 2.0 includes all surface water for smaller DWTPs (3,911 surface water intakes at 2,570 DWTPs) versus large water systems (2,915 intakes at 1,377 DWTPs) to compare the impacts of DFR to source water at those DWTPs. The research hypothesis was rejected as there was likely no statistically difference between the levels of DFR in surface water for smaller versus larger DWTPs (t-test, $t(2917) = -0.536$, $p = 0.274$). Smaller DWTP intakes located on lower Strahler stream orders were predicted to have a higher frequency of DFR than those for larger water systems. Low order streams are frequently dependent on treated wastewater effluents and vulnerable to high variation in the level of de facto reuse under mild droughts (7Q10 or Q95 low flow). The study investigated the current employment of advanced unit processes (i.e., activated carbon, ozonation, ultraviolet, nanofiltration) at DWTPs to evaluate the capability of removing CECs from source water. Smaller DWTPs lack a doubled number of advanced technologies compared with larger systems. There is no installation of any advanced unit processes at 52% and 23% for smaller versus larger DWTPs, respectively. Thus, when de facto reuse occurs, plus the lack of advanced technology at DWTPs can pose higher risks to CECs to the communities they serve. As larger DWTPs frequently supply surface water to populated communities, the total exposed population to the risks of CECs in source water could be extremely significant at larger DWTPs than smaller systems. The model estimated at DFR >1%, this could pose risks to a population served by larger DWTPs as 40 times as those served by smaller systems with 71 million and 1.7 million people, respectively. The model estimated a total of 73 million people relying on surface water impacted by DFR >1%. When predicted DFR was > 10%, the total exposed population to the risks of CECs to source water was estimated as 12.3 million people. Future studies can use DFR results to conduct epidemiological and risk assessment studies for communities impacted by wastewater derived CECs and identify communities that would benefit from advanced treatment processes that remove CECs.

In conclusion, the spatially explicit DRINCS model has conducted a systematic analysis of the extent of de facto potable reuse in source water across the United States. The development of the DRINCS model version 2.0 facilitates understanding of DFR in the extent of attenuation of contaminants and retention time in SW which can mitigate the public health risks to CECs. Quantification of DFR results can be used in epidemiological and risk assessment studies to understand and support direct potable reuse schemes in augmenting advanced treated reclaimed water with potable water supply.

The federal Clean Water Act (CWA) established the treated effluent discharges criteria to eliminate pollution and ensure the nation's waters to be "fishable and swimmable", but the regulated water quality limits do not reflect the drinking water standards. The Safe Drinking Water Act (SDWA) protects the public health by using risk factors (10^{-6} for chronic illness and 10^{-4} for acute (microbial) illness. Neither the CWA nor the SDWA regulates all potentially wastewater-derive contaminants which can pose health risks to the public. Decision-makers, water plant managers, and the population they represent must be notified of the relevant health issues concerning water quality and may be aware of the occurrence of de facto reuse in surface water supplies to improve water quality and achieve sustainable service,. As investment in advanced treatment technology may be costly and slow in coming for small community water systems impacted by DFR, it is possible to consider some quick and cost-effective solutions such as properly managed decentralized water production (e.g., point-of-entry, point-of-use treatment unit, and/or bottled water). Advanced treatment installed at DWTPs and increased monitoring of small Strahler stream orders (Order 1 and 2) associated with the highly impacted by DFR could be considered within the SDWA to protect the source water quality and public health. The adjustment could consider that smaller utilities may be impacted more due to financial trains or may have fewer resources to install advanced treatment. There is a need for a warrant policy for smaller utilities such as lower standards to control source water or supporting grants to upgrade the water plants. Findings from the DRINCS model could facilitate the development of contamination prediction tools or research on monitoring of CECs in surface water to understand and mitigate the risks to public health.

Recommendations for Future Work

It is essential for decision-makers, water plant managers and the population they represent to be aware of the relevant health issues concerning the impacts of de facto reuse on drinking water sources to improve water quality and achieve sustainable service. Findings from the DRINCS model could facilitate the development of contamination prediction tools or research on monitoring of CECs in surface water to understand and mitigate the risks to public health. Possibly, the DRINCS can be integrated within the Qualitative Microbial Risk Assessments (QMRA) framework to identify potential risks to human health. Optimization models and DRINCS can support decision making in retrofitting wastewater treatment plants or promoting new construction of water reclamation facilities. Future studies can extend the DRINCS model capability and functions to predict concentration in potable water, or evaluate exposures to CECs, and links to potential health risks. This could be done by creating new Python programmed tools within the ArcGIS framework, so they can be added to the current model tool.

Additional efforts could be made on the update DRINCS periodically to provide decision-makers with a better understanding of the extent of de facto reuse in the nations' water potable supply. For example, CWNS survey comes every four years or SDWIS updates the data on violations and enforcement every quarter of the year. to 10 years).

Advanced treatment and increased monitoring of surface water sources associated with the occurrence of de facto reuse could be considered within the SDWA to protect the public health and ensure safe drinking water. The future application of the DRINCS model can be a web application or a stand-alone software, so it can be easy to visualize the data and share the tool with other users. Many ArcGIS applications, such as ArcGIS

Dashboard or StoryMaps get inspiration from DRINCS model to create an educational tool in drinking water quality and potable reuse for kids between 8 to 12 years old. The DRINCS model can be further developed by integrating the GIS database connected with a real-time sensor, for example, to consider the variation in effluent discharge flows due to seasonal changes or plant operation hours. Other considerations could be considered additional point sources or non-point sources such as combined sewer overflows, industrial spills, hospital effluents, or agricultural runoffs, etc in the quantification of de facto reuse in potable supply and evaluating the status of surface water source protection. Stormwater or industrial treated effluents may contain a broader range of contaminants into surface water than municipal WWTP treated effluents. Agricultural runoff is considered as a non-point source of contaminants into surface water which can contribute significantly to the volume of effluents. The risks of DFR are associated with contaminants of emerging concerns in surface water supplies. They include wastewater-derived contaminants, transformed contaminants in surface water, trace organic contaminants, pathogens, disinfection by-products (regulated or unregulated), etc. It is important to evaluate the contribution of treated wastewater into source water, the level of treatment at a WWTP, unit processes installed at DWTP downstream, and the proximity analysis (travel time and proximal distance) and other factors (climatic condition, depth of the river, geology, and temperature).

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APPENDIX A

FOOD ENERGY WATER ANALYSIS AT SPATIAL SCALES FOR DISTRICTS IN THE YANGTZE RIVER BASIN (CHINA)

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Abstract

Understanding the nexus between food, energy, and water (FEW) systems is emerging as a critical area of study since federal research agencies in North America and Europe began highlighting needs related to data collection/management, systems optimization, and opportunities for new technologies. Little information regarding FEW systems exists across Asia, including within the Yangtze River basin, despite having 1/15th of the world's population living within the basin and generating as much as 40% of the Chinese gross domestic product. This research provides a case study of a FEW systems with analysis in the Yangtze River basin, showing the spatial and temporal variations in water availability/use, food production, and energy production. At a district-level scale in China, we integrated key Chinese datasets from multiple industrial, commercial and agricultural sectors together with key land use and hydrological information to evaluate the FEW parameters normalized to the land area of each district

rather than the commonly used approach where FEW consumptive parameters are normalized to population (i.e., per capita). The results illustrated the types of datasets currently available within China to conduct FEW system analyses and identified districts that are net producers or dependents regarding food, energy, or water. In the northeastern portion of the Yangtze River basin have several districts that are net negative relative to the amount of water that falls within the district boundaries versus all water use plus evaporation, with the most stressed districts lacking as much as 0.5 to 1 meter annually of equivalent rainfall per unit land area. The geospatial analysis concludes that policies to manage the FEW systems cannot be considered for a single district alone, nor the Yangtze River watershed in its entirety, but instead needs to consider the interdependencies among districts and consider encouraging growth (agriculture, industry, or population) within more water-abundant regions.

Keywords: *climate; food–energy–water system; Yangtze River basin*

Introduction

Many complexes, interrelated problems facing human society today relate to the production, distribution, and use of food, energy, and water production, distribution, and use, especially in developing countries (Bazilian et al. 2011). Food-energy-water (FEW) systems have inherent antagonisms, and the development of one sector usually depletes resources in the two other sectors (Chang et al. 2016b). An emerging body of research on the FEW nexus in Europe (Bhaduri et al. 2015, Hang et al. 2016) and the United States of America (Mortensen et al. 2016, Smidt et al. 2016) is beginning to break down barriers between different institutions (e.g., separate government ministries) to inform coordinated decision making about FEW systems (Ferroukhi et al. 2015). There is a recognized need to improve data aggregation and visualization techniques to facilitate coordinating policies in a holistic manner towards sustainability goals that consider interconnected FEW systems (Chang et al. 2016a).

Compared with abundant research on FEW systems at global (D'Odorico et al., 2018), European or North American scales, relatively few studies directly quantifying case studies of FEW systems are available for China despite China having the world's largest population and a rapidly developing economy that is applying stress to FEW systems (Jiang 2015). The Yangtze River (called as Chang Jiang in China) is the longest river in Asia and the third-longest in the world. The river's basin is home to more than 400 million people (i.e., 1/15th of the world's population). This basin alone would be the third most populated country in the world. The Yangtze River flows for 6,300 kilometers from the glaciers on the Qinghai-Tibet Plateau in Qinghai eastward across southwest, central and eastern China before emptying into the East China Sea at Shanghai. The basin represents 20% of the land area of the People's Republic of China, is home to 30% of the country's population, and conveys 30% of the water flow within China (Zhang et al. 2006) (Varis and Vakkilainen 2001). We focused on the Yangtze River basin because of its large role in the culture and economy of China. The Yangtze River basin generates as much as 40% of China's gross domestic product (Chen et al. 2014). Despite these staggering statistics, there is no published integrated FEW system analysis for the Yangtze River Basin. This limits the ability of the country and, because of its scale, the world to understand the holistic FEW system management.

A major gap in understanding and managing the global FEW systems will require knowing where local freshwater availability is sufficient to sustain future water needs

(D'Odorico et al., 2018). At a very localized scale for a major city in China (Beijing), an analysis of "virtual water" showed that whereas local water supplies, including reuse of wastewater, was adequate to meet industrial and environmental water uses within the city, the population needed to import grains and livestock, which had a water footprint from outside the city (Ye et al., 2018). In most cases for cities across developed countries, the water footprint is dominated by virtual water embedded in food (Chini, Konar, & Stillwell, 2017). Recognizing the dominant water footprint of the food system has been crucial in identifying strategies (e.g., irrigation practices, dietary changes) can be applied to meet food or water security and sustainability goals (Davis et al., 2016). One study suggests that because of the high water demand of food systems that agricultural water use efficiency could free up enough water for growing urban use in 80% of high-conflict watersheds around the world, including several regions in the Yangtze River where identified where modest (<5%) irrigation efficiencies could help overcome surface water deficits (Flörke, Schneider, & McDonald, 2018). The average water footprint for citizens in China (1,071 m3/year) was 60% lower than in the United States, and in both countries, this was dominated by virtual water associated with food consumption (Hoekstra & Mekonnen, 2012). Changing diets in China may lead to increases in this water footprint. Climate change has and will impact the Yangtze River basin in China (Han, Xu, Yang, & Deng, 2015), and it is likely that extreme hydrological events (e.g., floods, droughts) will increase in frequency, while annual mean streamflow will likely be remained constant (Yu, Gu, Wang, Xia, & Lu, 2018). Thus, regions within the Yangtze River watershed will see shifting climate patterns in local water availability for food, municipal, and energy production needs, which will necessitate developing a framework for managing real and virtual water within the basin (Rasul & Sharma, 2016). While general global trends and mega-city specific trends have been well studied, there is a lack of understanding at a regional (i.e., district) scale on water availability and FEW systems throughout the Yangtze River watershed.

This paper quantifies and compares components of the food, water, and energy system spatially, at the district level, within the Yangtze River basin. Food production, energy production, and water availability/use is normalized to land area (km2), rather than the more frequently applied normalization to the population when consumption or use of FEW resources are evaluated because generation of FEW resources occurs at a spatial or landscape scale. For select cases, population normalized FEW consumption values are also provided and discussed relative to other literature values. The objective of this study was to identify how various districts in the Yangtze River contribute differently, and show interdependence upon, to the production and consumption of food, water, and energy. Using an ArcGIS data management approach, we integrated datasets from multiple governmental agencies and applied ArcGIS models to 1) analyze geospatial patterns in land use and land cover (LULC), 2) conduct a preliminary water balance based on precipitation, potential evaporation, and water intake of each district, and 3) analyze spatial distribution characteristics of FEW components across agricultural, industrial and municipal sectors. This study was the first FEW analyses across the Yangtze River basin, and it focused on identifying and spatially quantifying key drivers for the FEW systems in this basin.

Data Sources and Methodologies

Study Site. Watershed boundaries and important cities in the Yangtze River Basin are illustrated in Figure SI.1. Figure SI.1 illustrates the river course and watershed boundaries, and the Yangtze River watershed can be divided into upper, middle and lower reaches based on landscape and climatic characteristics. The highest elevation point in the Yangtze River basin is 6621 m, located near Geladaindong Peak. From the river origins to Yichang, where Three Gorges dam located, is called the upper reach. In this reach, the river flows from high elevation plateaus and mountains and into the fertile valleys near the bottom of the reach. The middle reach, which spans from Yichang to Poyang Lake, receives the most precipitation (see below) and accounts for 40% of the river basin area. Here the Yangtze River gradient decreases and its course meanders, forming a broad river and slow stream flows. The lower reach, which starts at Poyang Lake, has flat terrain, short tributaries, stable flow and a dense water network, ending at the estuary in the East China Sea.

Data Sources & Computational Methodologies. ArcGIS was used to aggregate data from different publicly-available sources (see Supplemental Information). Geographic Information System (GIS) layers for topography as well as stream networks and district boundaries were obtained from China Geological Survey (NGCC 2018). The land use and land cover dataset were provided by Cold and Arid Regions Sciences Data Center at Lanzhou (Ran et al. 2010). The primary dataset included the quantity of water intake and water consumption collected from water resources bulletins and the streamflow of rivers in Yangtze River basin (China 2005-2016a, NGCC 2018). All provinces in China publish water resource bulletins annually that include macro data about water consumption and wastewater production within cities (China 2005-2016a).

They also publish annually economic and social development bulletins and energy production and consumption bulletins (China 2005-2016b). The output of grain (cereals, beans and potatoes) dataset was collected from economic and social development bulletins (China 2005-2016b). All data were based on the prefecture-level administrative regionalization. Additional information was obtained from literature (Albala-Bertrand 2016, Amadei et al. 2013, Besha 2011, Chang et al. 2003, Jiang 2015, Yang et al. 2016, Zheng et al. 2014)

Historical streamflow data collected from 1954 to 2014 were obtained from Yangtze River Water Resources Committee (China 2005-2016a). Table SI.1 shows average annual stream flows at twelve hydrological stations along the Yangtze River (shown in Figure SI.1). On annual average, 26,121 m^3/s leaves the middle reach and 26,752 m^3/s leaves lower reach before entering the East China Sea.

The annual precipitation and potential evapotranspiration data were derived from high-resolution gridded datasets provided by climatic research unit, and potential evapotranspiration was calculated from a variant of the Penman-Monteith formula (Harris et al. 2014). Within different districts (i) of the Yangtze River a district level natural climatic water balance (CWB_i, mm) across the land area of the basin was computed as a function of measured annual precipitation (P_i, mm) minus calculated annual evapotranspiration (E_i, mm):

$$CWB_i = P_i - E_i \qquad \text{Equation 1}$$

The total water balance (TWB_i) the Yangtze River a district across the land area of the basin was computed as a function of P_i (mm) minus E_i (mm) minus both industrial water intake (*IWI*, mm) and residential water intake (*RWI*, mm) normalized to district (*i*) land area (m^3/km^2):

$$TWB_i = P_i – E_i - IWI_i - RWI_i \qquad \text{Equation 2}$$

Water consumption for electrical energy production was calculated. In 2010, 74% of the power in China was produced by thermal-electric power plants that consumed 2.45kg/kWh of produced power (沈旭 et al. 2013); reported values decreased from 3.00 to 2.45 and 2.30 kg/kWh between 2005, 2010 and 2015, respectively. Power production information was obtained from energy production and consumption bulletins of each involved province.

Population & Land Use Distributions. Figure 1 shows the geospatial distribution of land use (forest, grasslands, croplands, urban and build areas and waterbodies (Ran et al. 2012)) and population with the Yangtze River basin. The upper reach, except within the Sichuan basin, is characterized by higher elevations, lower average temperatures, mostly grass and forest lands and low population density. The Sichuan basin is a lowland region in upper reaches of Yangtze River basin, and it is heavily populated with more than 100 million inhabitants. The relatively flat lands and fertile soil in the upper reach support extensive irrigated croplands. Two dense urban areas in the upper reach are Chengdu and Chongqing, which are the major economic centers in southwest China.

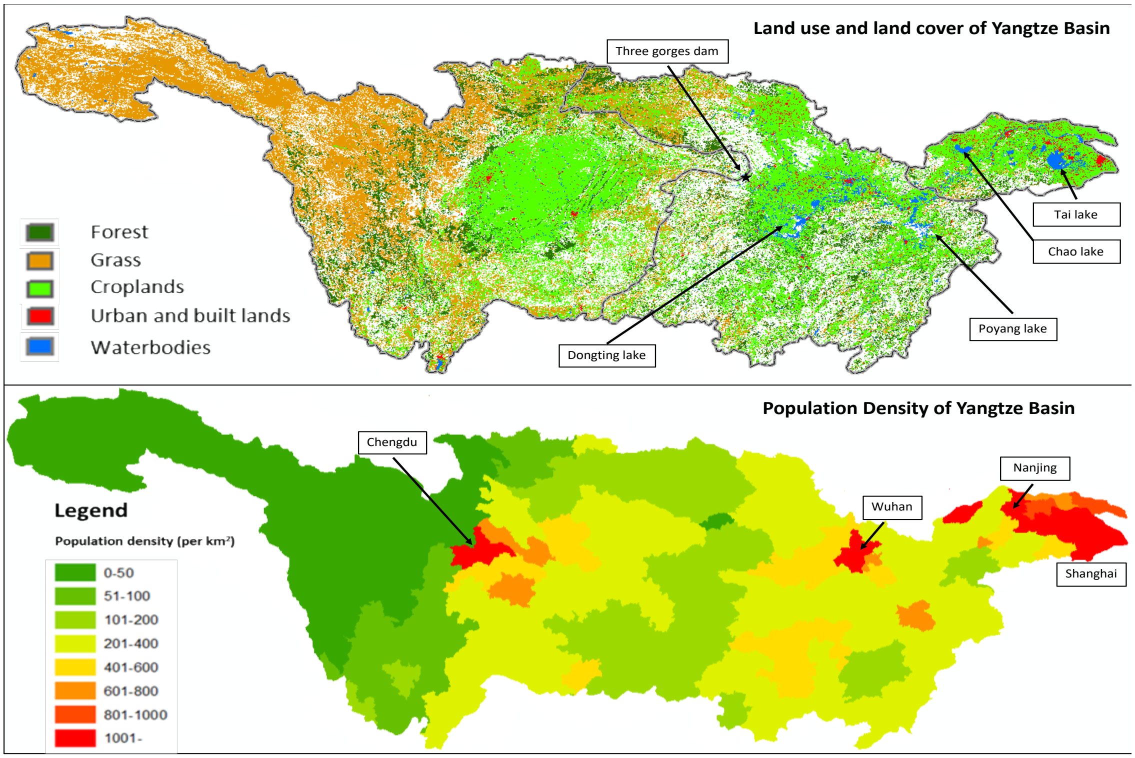

Figure 1 Map of land use and land cover and population density of the Yangtze River basin. Tributary basins, lakes and Three Gorges dam are shown on the top map. Cities are shown on the bottom map for 2010

Broad plains formed of alluvial deposits crisscross the middle reach of the basin. This area is known as China's major granary and is characterized by moderate temperatures and abundant rainfall that support large areas of non-irrigated croplands. Population in major cities such as Wuhan and lands across the basin have seen more significant urbanization than the upper reaches, resulting in a loss of natural grass and forest lands.

The Yangtze River delta in the lower reach includes the economic centers of Shanghai and Nanjing and is the most affluent region in China. In the lower reach, the Yangtze River widens as the land gets flatter, and streamflow from the upper basin combined with fertile soil make the Yangtze River delta suitable for growing rice. Over the past few decades, the population and level of urbanization has increased dramatically in select parts of the basin.

Results and Discussions

District-Level Water Balance. Average annual precipitation across the Yangtze River basin ranges from 400–2000 mm (Figure 2). Generally, lands located south of the Yangtze River have more precipitation than regions located north of the river in middle and lower reaches. The middle reach and coastal lands receive the most precipitation. Mountains northwest of Ya'an influence monsoon-related rainfall when humid southerly winds blow against the mountains, lifting moisture and enhancing rainfall. This is one example of many areas across the Yangtze basin where annual rainfall amounts don't always reflect constant rainfall patterns throughout the year.

Using annual estimates for precipitation and evapotranspiration within each district and without considering water uses or in-stream flows, a macro-scale water balance was conducted to identify districts with a net excess or deficiency of water simply based upon hydrologic inputs (rainfall) and outputs (evaporation) (i.e., Equation 1). Figure 2 shows that approximately half the basin is a net producer of water (i.e., over 250 mm more annual rainfall than evapotranspiration) and shows that the distribution over the basin is highly spatial. Individual districts that are net producers (positive values in Figure 2) or have a net deficit (negative values in Figure 2) is neither "good" nor "bad" but begins to show the interdependencies for water availability among and between districts scattered throughout the basin.

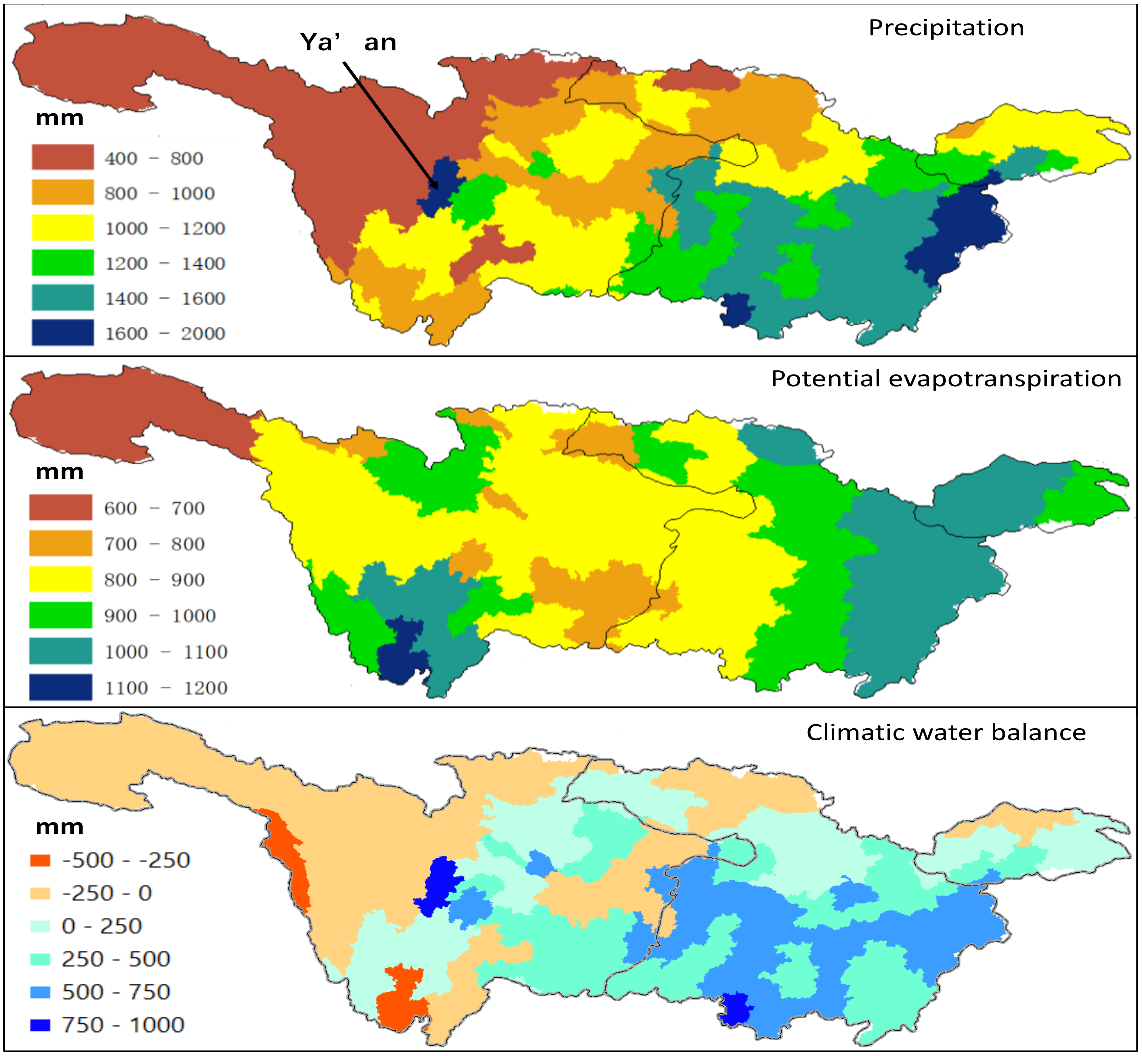

Figure 2 Spatial distribution across districts for 2010 in the Yangtze River basin of precipitation, calculated potential evapotranspiration and calculated climatic water balance (Equation 1).

Because of irregular precipitation throughout the year and snowfall at higher elevations, this simple macro-scale water balance provides only minimal information on the availability of water at the right time and place within the basin. Precipitation runoff and snowmelt generate streamflow into the tributaries and mainstem of the Yangtze River (Table SI.1). Based upon historical data, the annual average streamflow leaving the middle reach (26,121 m^3/s) is roughly equivalent to the streamflow out of the lower reach

(26,752 m^3/s), which enters the East China Sea, suggesting the lower basin may be approaching a steady state intake of river water, discharge of municipal, industrial or agricultural wastewater, with some contribution of stormwater runoff. Annual differences in streamflow exist and are described elsewhere (Wang et al. 2017c). Later we describe the impact on water balances of additional demands for industrial, municipal and agricultural water. However, it suggests the lower region may be nearing a tipping point, where it becomes a net consumer of river water.

Human Water Uses. In China, water use data is divided into three sectors: agricultural (agriculture, forestry, animal husbandry and fishing), industrial (mining, manufacturing and power) and municipal (service industry, information technology, education and living). Available water data were used from 2003 through 2014 annual bulletins. Three years (2005, 2010 and 2014) were selected to illustrate the effect of rapid urbanization and industrialization in China on spatial water uses. To enable spatial comparisons, data across each district was normalized by the land area, and Figure 3 shows spatial water intake patterns for these three years for each sector. The total municipal water intake volume for the entire Yangtze River basin increased from 22.54 billion m^3 in 2005 to 32.39 billion m^3 in 2010 and 32.82 billion m^3 in 2014. In most districts, the municipal water use is low compared with agricultural or industrial intakes. There are many districts in the upper and middle reaches with less than 50 m^3 water per km^2. Densely populated districts intake 100 to over 200 m^3 water per km^2. This includes districts of Chongqing, Wuhan and Shanghai, which intake 1.9, 1.0, and 2.4 billion m^3 water, respectively.

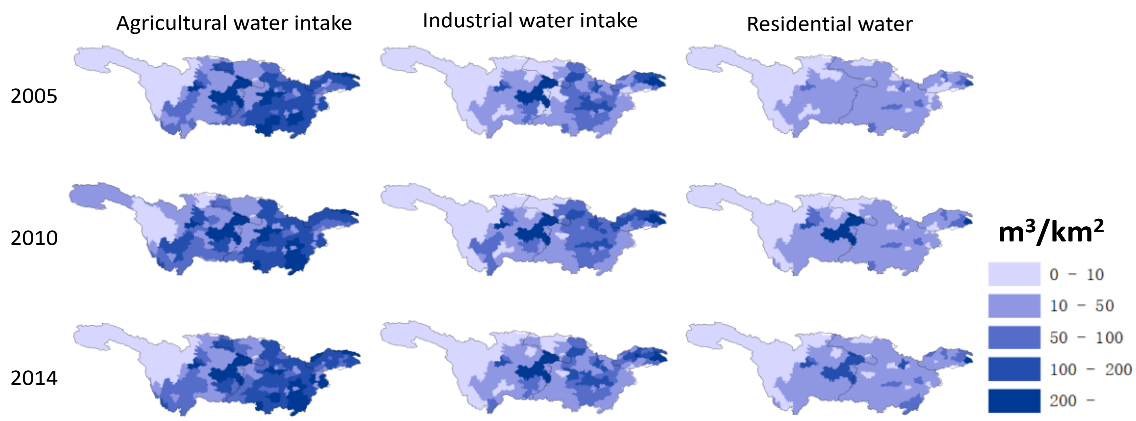

Figure 3 Land area based annual water intake (m^3/km^2) of agricultural, industrial, and municipal sectors in 2005, 2010, and 2014.

In the upper reach, the water intake for the agricultural sector, industrial sector, and the municipal sector is 53%, 35%, and 12% respectively. In the middle reach, the percentages are 61%, 32%, and 7%, respectively. In the lower reach, the percentages are 42%, 50%, and 8%, respectively. Thus, agricultural sectors intake the majority of water in the upper and middle reaches, while industry accounts for the majority in the lower reach. The municipal sector accounts for a relatively small percentage (7% to 12%) of water intakes across the entire Yangtze River basin; however, the percentage increases in high population density districts (e.g., Chengdu, Wuhan, Nanjing, and Shanghai). For example, municipal water accounts for 32% of the total water intake in the district of Shanghai, compared against agricultural (18%) and industrial water (50%) use.

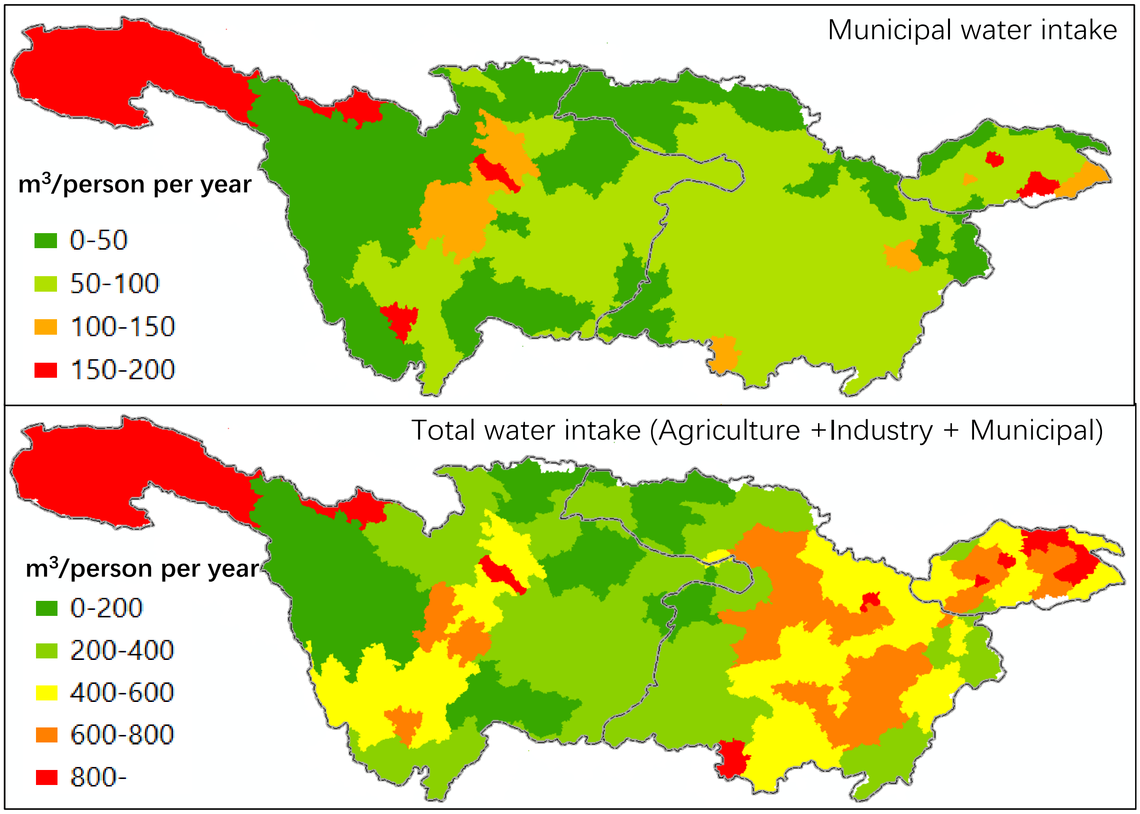

Figure 4 Population based water consumption (m^3/person per year) in each district for municipal water intake only and cumulative water intake (Municipal + Agriculture + Industry (m^3)) for each district in 2010

Not all water intakes result in consumptive use, and a fraction of the water returns to the river. Separately, we describe how relationships between population, municipal water intake and construction of sewage treatment plants contribute to streamflow along the Yangtze River (Wang et al. 2017c), including the following two key conclusions relevant for FEW systems: 1) municipal wastewater produced in the Yangtze River basin increased by 41% between 1998 and 2014—from 2580 m^3/s to 3646 m^3/s—in conjunction with China's investment in public infrastructure; and 2) under low flow conditions in the Yangtze River near Shanghai, treated wastewater contributions to river flows increased from 8% to 14% between 1998 and 2014. Figure 4 shows additional insights when municipal water consumption and total water intake is normalized to population. Municipal water consumption varies little across the basin, whereas the cumulative water consumption rates (industry, agriculture plus municipal) are higher on the fringes of densely populated urban areas and in the southeastern portion of the Yangtze River basin (Figure 4). This illustrates the importance of adjoining districts on supporting the prosperity of higher density population districts.

Food Production and Agricultural Water Use. The Yangtze River basin produces a diverse array of agricultural products (Liu et al. 2014), with the major group being grains, beans and potatoes. Figure 1 shows the spatial distribution of croplands, and Figure 40 shows the district level areal production of grain (tons/km^2), which totaled 205 million tons in 2014 (Figure SI.3 shows population normalize energy production). Grain production per unit area is lower in the upland parts of the upper reach (i.e., <100 tons/km2) and gradually increases throughout the middle and lower basins. Sichuan Basin is the main grain producer in upper reach, and the district of Chongqing has the highest annual total regional grain output (11.5 million ton). Abundant croplands in the middle reach account for 98.4 million tons of annual grain, with the highest output coming from the Han River basin. The lower reaches produce 35.9 million tons of annual grain, with the highest output (5 million tons) coming from the district of Nantong which is located near the estuary of the Yangtze River.

The basin's high grain output exerts a high-water demand. The agricultural sector water intakes in 2014 for the districts of Chongqing, Xiangyang and Nantong were 2.4 billion m^3, 2.4 billion m^3 and 2.2 billion m^3, respectively. This equates to 209 m^3 of water per ton of grain in the Chongqing district, 440 m^3/ton in Nantong and 480 m^3/ton in

Xiangyang. Between 2005, 2010 and 2014, there was a slight increase in the agricultural sector water intake from 104 billion m^3 in 2005 to 113 billion and 114 billion m^3 in 2010 and 2014, respectively. Agricultural water intake (Figure 3) correlates wells with grain output (Figure 5) throughout the basin.

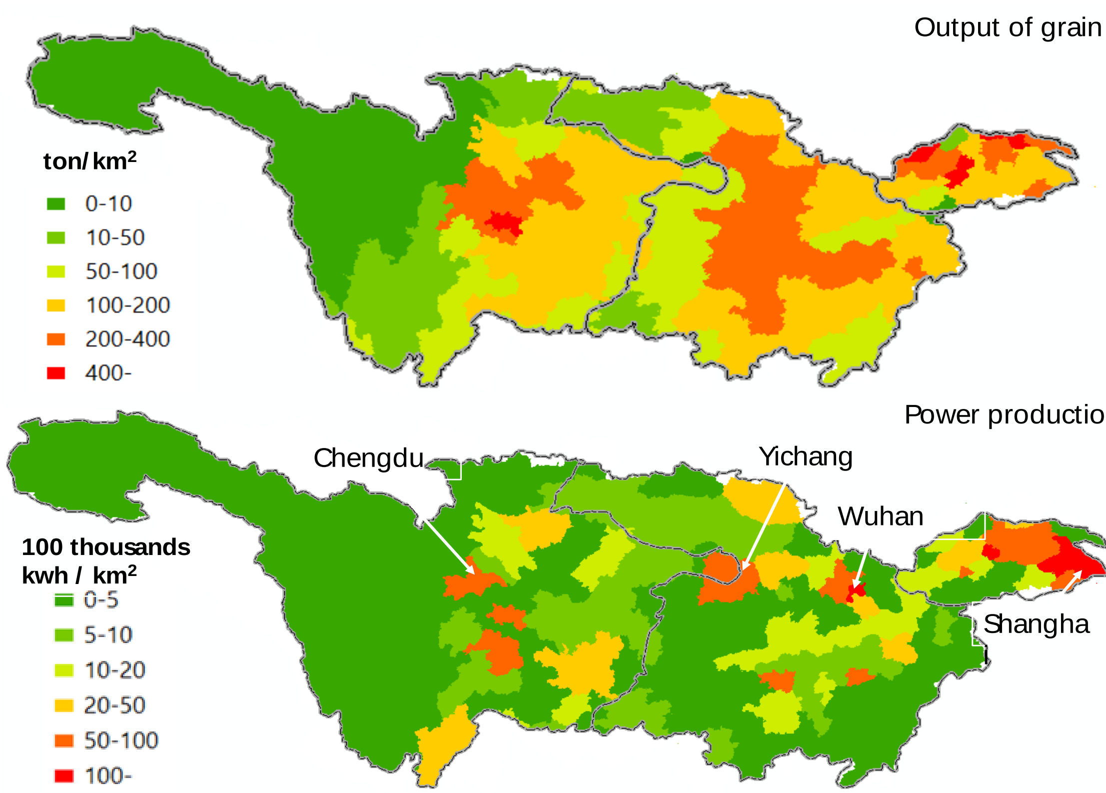

Figure 5 Land area-based outputs of grain (food) and power production (energy) from each district in the Yangtze River basin in 2010.

Energy Production and Industrial Water Use. Industrial water accounted for 66 billion m^3 in 2005, and 82 and 65 billion m^3 in 2010 and 2014, respectively. As illustrated in Figure 3 industrial sectors intake less water than agricultural sectors. Intense water intake districts are geographically spotted across the basin, including Chengdu, Chongqing and Wuhan in the upper and middle reaches and Changzhou, Wuxi, Suzhou and Shanghai districts in the lower reaches, where the industrial sector water intake in

2014 was 1.3 billion m^3, 2.0 billion m^3, 3.1 billion m^3 and 3.9 billion m^3, respectively. These intakes account for 48%, 56%, 57% and 50% of the total water intakes in these four districts of the lower reach.

Energy production facilities (coal, gas, nuclear and hydro power) distributed across different districts (Figure SI.2) account for a large portion of the industrial sector water intake. While hydropower generation may slightly increase evaporation from lake surfaces, the nuclear, coal and gas energy production facilities require large volumes of water for thermo-electric cooling towers. While once-through cooled power facilities may only evaporate on the order of 0.5% of the intake water, those with cooling towers evaporate on the order of 50% of the intake water (Sanders et al. 2014). Approximately 900 energy generation facilities are capable of cumulatively producing 1826 billion kWh. To contrast agricultural versus power generation outputs from each district, Figure 5 normalizes both of these outputs to land area (Figure SI.4 shows population normalize energy production). The energy production within each district (kWh/km2) utilizes water from within the district but can deliver energy to multiple districts. Compared against agricultural grain production which consumes water across larger land areas, point sources of water consumption for power production (Figure SI.2) are concentrated in fewer districts.

Power generating facilities are spatially located based upon availability of coal, gas or hydropower and near major population centers (Figures 1 and SI.2). The Three Gorges hydropower facility located near Yichang generates more than 160 billion kWh per year. Smelting facilities in districts with significant metal mining activities rely, in part, on local coal powered facilities to produce electricity, resulting in several districts with relatively small populations having relatively high energy production capacity. Water intake for power production is greater in 2010 (Figure 3) than 2014. This corresponds a decline in the growth rate of the industrial output from China. Production and rapid adoption of photovoltaic and thermal solar power is occurring across China (Wang et al. 2017a, Wang et al. 2017b), and the water footprint of solar power in China was recently suggested to be higher than previously thought and may account for >30% of the total industrial water use in some major cities (Wu and Chen 2017).

The Food-Energy-Water nexus in The Yangtze River Basin. Grain output, power output and total water intake of each district in 2014 were used to study the FEW nexus of the Yangtze River basin. Figure 6 shows the FEW systems net water interdependency of each district in the basin (i.e., precipitation, evaporation and all water intakes), represented as net mm of water across the landscape (Equation 2). Net negative values indicate districts more strongly dependent upon river flow from up-river locations, while net positive values indicate a surplus that flows to down-river locations. Districts close to net zero cumulative water, or negative, are water stressed regions that depend on water management policies in the upper watershed, including south to north water transfers from Three Gorges Dam (Figure 1) (Zhao et al. 2015). In the northeastern portion of the Yangtze River basin have several districts that are net negative regarding cumulative water, with the most stressed districts lacking as much as 0.5 to 1 meter annually of equivalent rainfall per unit area.

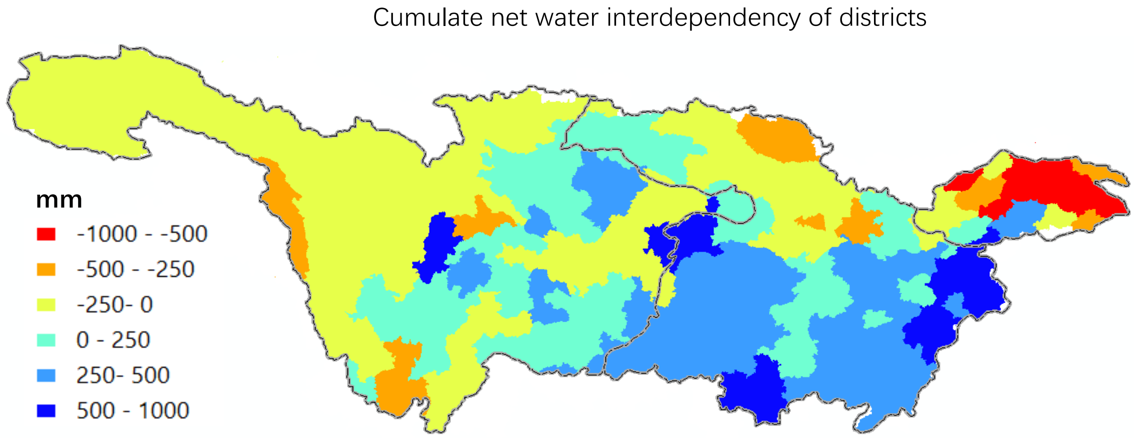

Figure 6. Net water intake by people across the Yangtze River basin in 2010 (based upon Equation 2).

Ultimately communities rely upon water for food and energy, and an interesting juxtaposition of the FEW is presented in Figure 4 for the total water intake per district normalized to population within the district. While annual municipal water consumption rates range up to 200 m^3/person/year, accounting for agriculture and industrial withdraws raises these cumulative rates to 800 m^3/person/year. Municipal water consumption varies little across the basin, whereas the cumulative water consumption rates are higher on the fringes of densely populated urban areas and in the southeastern portion of the Yangtze River basin where the largest amounts of net positive cumulative water (Figure 6) is located; Figure SI.5 shows similar patterns net water consumption or production from across the district (m^2) using the mm of water data from Figure 6 and then normalizing values to population within each district. This case study demonstrates the value of data visualization to consider spatial district level and temporal analysis of FEW systems to maximize available resources within districts across the Yangtze River basin.

Conclusions

This case study on food-water-energy systems across one of the largest watersheds in the world provides valuable insights into data availability in developing countries and geospatial patterns. Municipal water consumption varies little across the basin, despite very different rainfall patterns. This places a high level of reliance upon water in the Yangtze River, and highlights the reliance upon large population centers at the downstream end of the basin on generally less populated but more agriculture and mining intensive activities in upstream regions of the basin. While municipal water consumption rates range up to 200 m^3/person/year, accounting for agriculture and industrial withdraws raises by nearly 4X the cumulate water usage per capita within certain districts. Agricultural water intake correlates geospatially well with grain output (Figure 5) throughout the basin. Thus, as agricultural activity increases to provide food to growing populations, either more water will be withdrawn from the basin upstream or technological advances in agricultural water efficiency are needed to prevent droughts and challenges to large downstream municipalities. We observed a decline in water intakes for power production between 2010 and 2014, corresponding with a decline in the growth rate of the industrial output from China but also with increased reliance upon solar-based energy production (Albala-Bertrand 2016, Besha 2011, Yang et al. 2016). As China adopts more aggressive renewable energy production (Song and Wang 2018), it may decrease water use by coal-fired power generating stations and increase water availability for agriculture or municipal uses.

Many of the most water stressed districts are in the northeastern portion of the Yangtze River basin and much of the water demand is for municipal rather than industrial or agricultural use. Consequently, with their proximity to ocean – the large cities in this area may be candidates to consider ocean desalination projects, where generated municipal wastewater could be used to supply water to the local agriculture or industry water users. Desalination has an embedded energy density of roughly <1 kWh/m^3 for treatment (Qin et al. 2019, Werber et al. 2017), and can be equally as energy intensive to pump treated water inland. Thus, while desalination may address water demand challenges there will be an associated water demand somewhere in the basin associated with the energy required to desalinate and transport seawater.

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APPENDIX B

QUANTITATIVE DETECTIONS ASSESSMENT

Location With Max Detection	Analyte	CAS Number	n	Qualitative Frequency of Detection %[a]	Quantitative Frequency of Detection %[a]	Max Source Water Conc ng/L[a]	Max WWTP-Influenced Conc ng/L[b]	Source as % of WWTP Impacted Conc
DWTP 02	Tri (2-butoxyethyl) phosphate	78-51-3	25	36	4	470	—	—
	Tri(2-chloroethyl) phosphate	115-96-8	25	32	4	65	—	—
DWTP 03	Tramadol	27203-92-5	25	32	16	1,723.00	1,311.30	1.8
	Trimethoprim	738-70-5	25	28	16	9.9	198.8	5
	Diltiazem	42399-41-7	25	20	8	15.5	56	27.7
	Ibuprofen	15687-27-1	25	8	8	17.7	1,620.00	1.1
	Furosemide	54-31-9	25	4	4	17.5	—	—
	Paraxanthine	611-59-6	25	4	4	29.2	—	—
DWTP 04	Galaxolide (HHCB)	1222-05-5	25	36	36	110	1,400.00	7.9
	Metoprolol	51384-51-1	25	52	32	37.8	367.1	10.3
	Carbamazepine	298-46-4	25	56	28	35.7	382.7	9.3
	Estrone	53-16-7	25	52	20	0.3	31.5	0.9
	Hydrochlorthiazide	58-93-5	25	24	20	67.3	—	—
	Desvenlafaxine	93413-62-8	25	28	16	60.4	1,953.50	3.1
	Bromoform	75-25-2	25	12	12	88	—	—
	Caffeine	58-08-2	25	32	12	2,790.90	1,275.90	7.1
	Triclosan	3380-34-5	25	52	12	3.5	534	0.7
	Valsartan	137862-53-4	25	20	12	79.2	—	—
	Cotinine	486-56-6	25	16	8	18.9	68.1	27.7
	Fexofenadine	83799-24-0	25	12	8	163.1	2,047.40	8
	Venlafaxine	93413-69-5	25	12	8	26.3	407.3	6.5
	Atenolol	29122-68-7	25	28	4	29.8	551.1	5.4
	Diphenhydramine	58-73-1	25	16	4	10.3	145.3	7.1
	Progesterone	57-83-0	25	12	4	0.1	0.9	16
DWTP 12	Bisphenol A	80-05-7	25	4	4	28.5	163	17.5
DWTP 18	Triclocarban	101-20-2	21	57	24	2.9	—	—
	Dihydrotestosterone	521-18-6	24	4	4	0.3	1.6	19.8
	N,N-Diethyl-meta- toluamide	134-62-3	25	48	4	98	—	—
DWTP 21	Testosterone	58-22-0	25	4	4	0.2	1.1	14.1
	Atrazine	1912-24-9	25	44	24	323.3	5,170.00	6.3
	Metolachlor	51218-45-2	25	36	12	130	1,490.00	8.7
	Norverapamil	67018-85-3	25	20	8	47.2	—	—

	10-hydroxy-amitriptyline	1159-82-6	25	4	4	0.3	9.4	3.2
	Amitriptyline	50-48-6	25	4	4	12.1	10.2	118.1
	Verapamil	52-53-9	25	20	4	45.9	12.9	355.7
DWTP 22	PFHpA	375-85-9	25	96	96	184	—	—
	PFHxA	307-24-4	25	96	96	55.1	—	—
	PFNA	375-95-1	25	96	96	41.4	—	—
	PFBA	375-22-4	25	92	92	96.8	—	—
	PFPeA	2706-90-3	25	92	92	501	—	—
	PFOS	1763-23-1	25	96	88	48.3	—	—
	PFOA	335-67-1	25	100	76	112	—	—
	PFDA	335-76-2	25	92	60	31.1	—	—
	PFUnDA	2058-94-8	25	36	32	2.9	—	—
	PFDoDA	307-55-1	25	20	8	0.3	—	—
	Carisoprodol	78-44-4	25	16	4	5	155.9	3.2
	Fluconazole	86386-73-4	25	8	4	33.7	232.4	14.5
	Meprobamate	57-53-4	25	32	4	14.2	405.9	3.5
	Sulfadimethoxine	122-11-2	25	8	4	7	1800	0.4
DWTP 24	PFBS	375-73-5	25	100	96	11.1	—	—
	PFHxS	355-46-4	25	92	92	44.8	—	—
	Tributyl phosphate	126-73-8	25	8	4	87	503	17.3
DWTP 26	Bupropion	34841-39-9	25	20	20	9.4	159.6	5.9
	Cholesterol	57-88-5	25	24	4	200	3170	6.3
DWTP 27	Methyl-1H-benzotriazole	136-85-6	25	48	44	1199.9	921	130.3
	Sulfamethoxazole	723-46-6	25	60	40	161.1	1500	10.7
	Pseudoephederine	90-82-4	25	24	20	4.5	89	5
	Desmethyldiltiazem	85100-17-0	25	8	8	6	19.8	30.3
	Methocarbamol	532-03-6	25	36	8	32.3	2627.3	1.2
	Hydrocodone	125-29-1	25	4	4	8.1	45.8	17.7
	Ranitidine	66357-35-5	25	4	4	13.1	313.9	4.2
DWTP 29	Lidocaine	137-58-6	25	20	8	29.7	408.8	7.3

[a]Source water concentrations are taken from Glassmeyer et al. 2017.
[b]Wastewater-influenced concentrations are taken from Bradley et al. 2017.
Conc—concentration, DWTP—drinking water treatment plant, WWTP—wastewater treatment plant

APPENDIX C

CALCULATED 7Q2 AT DIFFERENT STREAM ORDERS

	Streamflow,cfs		
	Percentile	Mountain creek	Navasota river
Stream Order 3	P1	0.000	0.000
	P5	0.000	0.000
	P10	0.000	0.000
	7Q2	0.000	0.000
	P20	0.000	0.000
	P25	0.000	0.030
	P30	0.000	0.150
	P40	0.100	0.510
	P50	0.570	1.100
	P60	1.700	3.100
	P70	4.500	9.900
	P75	7.425	17.000
	P80	12.000	29.000
	P90	48.000	102.000
	P95	115.000	402.000
	P99	947.980	2636.000
	Streamflow,cfs		
	Percentile	North Bosque R	San Marcos R
Stream Order 4	P1	0.000	64.000
	P5	0.300	81.000
	P10	1.400	93.000
	7Q2	2.150	127.000
	P20	4.100	117.000
	P25	6.600	131.000
	P30	8.900	149.000
	P40	15.000	181.000
	P50	24.000	211.000
	P60	39.000	261.000
	P70	73.000	339.000
	P75	103.000	397.000
	P80	143.000	473.800
	P90	336.000	720.400
	P95	685.000	1070.000
	P99	3511.300	3470.000
	Streamflow,cfs		
	Percentile	Leon river	Pecan Bayou
Stream Order 5	P1	0.000	0.000
	P5	1.700	0.000
	P10	5.100	0.000
	7Q2	3.379	0.000
	P20	12.000	0.200
	P25	17.000	0.300
	P30	22.000	0.400
	P40	36.000	1.000

	P50	68.000	2.100
	P60	166.000	3.700
	P70	358.000	7.400
	P75	482.000	15.000
	P80	697.000	46.000
	P90	1860.000	286.000
	P95	3300.000	379.300
	P99	6260.000	2500.000
Stream Order 6	Streamflow,cfs		
	Percentile	Sabine river	Colorado river
	P1	2.300	6.800
	P5	22.250	33.000
	P10	48.500	53.000
	7Q2	51.500	41.800
	P20	73.000	90.000
	P25	85.000	108.000
	P30	96.000	126.000
	P40	126.000	170.000
	P50	162.000	220.000
	P60	196.000	305.000
	P70	245.000	456.000
	P75	283.000	580.000
	P80	316.000	754.000
	P90	403.000	1670.000
	P95	456.000	3810.000
	P99	514.450	17235.00
Stream Order 7	Streamflow,cfs		
	Percentile	Brazos river	Nueces river
	P1	57.980	0.000
	P5	110.000	0.000
	P10	210.000	2.200
	7Q2	203.143	0.000
	P20	365.000	3.900
	P25	450.000	4.100
	P30	543.000	4.400
	P40	779.000	4.700
	P50	1060.000	5.000
	P60	1430.000	6.700
	P70	2020.000	23.000
	P75	2530.000	37.000
	P80	3250.000	77.000
	P90	6220.000	750.000
	P95	11755.000	3047.500
	P99	34000.000	12527.00
Stream Order 8	Streamflow, cfs		
	Percentile	Rio Grande	
	P1	464.620	
	P5	647.000	

	P10	748.000
	7Q2	728.000
	P20	871.800
	P25	937.500
	P30	1000.000
	P40	1118.000
	P50	1270.000
	P60	1420.000
	P70	1700.000
	P75	2025.000
	P80	2430.000
	P90	3678.000
	P95	4790.000
	P99	9095.000

APPENDIX D

ADDITIONAL INFORMATION ON DATA AND SOURCES

Table SI 1 Summary of additional sources of information used within this research

Provider	Description	Format	Source	Associated Figure
USEPA - Clean Watersheds Need Survey (CWNS 2008)	Locations of WWTP discharges in Texas (Latitude and Longitude)	Microsoft Access	https://ofmpub.epa.gov/apex/cwns2008/f?p=cwns2008:25:0:	Figure 1
Texas Environmental Commission Quality (TCEQ)- Texas Drinking Water Watch (TDWW)	Locations of DWTP intakes in Texas (Latitude and Longitude)	Single input	https://dww2.tceq.texas.gov/DWW/	Figure 2
	DWTP Unit Treatment Processes			Figure SI.1
US Geological Survey (USGS)- National Hydrography Dataset (NHD)Plus 2	US Stream Network (medium-resolution 1:100,000 scale)	Shapefile	http://www.horizon-systems.com/NHDPlus/NHDPlusV2_home.php	Figure SI.2
	US Hydrography Regions and Watersheds			Figure SI.3
US Geological Survey (USGS)	Hydrography (Major Rivers, Water Bodies, Watershed Boundary)	Shapefile	https://www.usgs.gov/core-science-systems/national-geospatial-program/national-map	Figure SI.7
Us Census Bureau	US Census, Cities, Counties and States	Shapefile	https://www.census.gov/programs-surveys/geography.html	Table SI.6
US EPA - Enforcement and Compliance History Online (ECHO)	US Annual Compliance Report Year 2015	Excel	https://echo.epa.gov/trends/comparative-maps-dashboards/state-water-dashboard	Table SI.7 Figure SI.8 Table SI.9
Environmental Flows Information System for Texas - USGS Hydrologic Information	Calculation of 7Q2 low flow index	Excel-based application		Figure 3

System Gauging Data US Geological Survey (USGS) National Hydrography Dataset (NHD)Plus 2	US NHD Stream gages	Shapefile	http://www.horizon-systems.com/NHDPlus/NHDPlusV2_home.php	Appendix C
ArcGIS Network Analyst tool	Proximity Analysis	Shapefile	ArcGIS version 10.4	Figure 4 and Figure 5
				Figure SI.4
				Table SI.2
				Table SI.2
Texas Drinking Water Watch (TDWW)	DWTP Unit Treatment Processes	Single input	https://dww2.tceq.texas.gov/DWW/	Figure 6 and Figure 7
US EPA - EPI Suite and Fate model LEV3EPIFugacity Model	Half-life of CEC surrogates	Application	https://www.epa.gov/tsca-screening-tools/download-epi-suitetm-estimation-program-interface-v411	Table SI.4 Figure SI.6